# JOURNAL
# OF
# GEOMAGNETISM
# AND
# GEOELECTRICITY

VOL. VIII NO. 2

SOCIETY
OF
TERRESTRIAL MAGNETISM AND ELECTRICITY
OF
JAPAN

JUNE 1956
KYOTO

JOURNAL OF GEOMAGNETISM AND GEOELECTRICITY



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The Editors

# Magnetic Interaction between Ferromagnetic Minerals Contained in Rocks (II)*

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(Read on 14th, May, 1956)

## Abstract

In this paper, the effect of magnetic interaction between coexisting minerals in the course of development of thermo-remanent magnetism (TRM) was examined. Various combinations of minerals in the ternary system $FeO$-$Fe_2O_3$-$TiO_2$ were derived by heat treating the natural ferromagnetic ilmenites. Coexisting phases produced by oxidation, reduction and phase transformations during heat treatments were found in parallel alternating structures. The main results were: (1) When both of the two phases are ferromagnetic, magnetic interaction acts negatively and the TRM of the composite system tends to be reverse TRM (RTRM). RTRM of the Haruna-type is the most typical example of this case, where the two phases are both ferromagnetic ilmenites. Intensification and artificial production of RTRM of this type were interpreted by the hypothesis of "partial exsolution" during the heat treatment at 600°C–700°C. Another type of RTRM where the two phases are titanomagnetite (Ti-Mt) and ferromagnetic ilmenite was produced by reduction. As for the case of combination of two Ti-Mts, the anomalous increase of TRM and the reversal of natural remanent magnetism, studied by other investigators, were referred to. (2) When non-ferromagnetic minerals, such as titan-hematite, rutile, non-ferromagnetic ilmenite or ulvöspinel, coexist with ferromagnetic ilmenite or Ti-Mt, the TRM and coercive force of the composite system are intensified remarkably: a linear relation $J_{tr}(T_0)/J_s(T_0) \propto H_c(T_0)$ was obtained for the composite of ferromagnetic ilmenite and titan-hematite and rutile.

As by-product of the present study, some information on the oxidation-reduction properties of ferromagnetic ilmenite was obtained: for example, oxidation was found to proceed in two distinct steps as the treatment temperature is raised.

## Introduction

Since the prediction [1] and the discovery [2], [3], of the RTRM (reverse thermo-remanent magnetism) of certain igneous rocks and its explanation in terms of magnetic interaction between the ferromagnetic minerals contained therein [4]–[7], the role played by the magnetic interaction during the process of development of remament magne-

* Contribution from Division of Geomagnetism and Geoelectricity, Geophysical Institute, Tokyo University. Series II, No. 64.

tism of rocks has been attracting the interest of a number of investigators [8]–[12]. The general interest shown in this matter is related to the extremely important nature of its function in the field of palaeomagnetism [3].

Through the study of the mechanism causing the Haruna-type RTRM [12], the magnetic interaction essential in that case was shown to be between two ferromagnetic solid solutions of the ilmenite-hematite (Il-Ht) series contained in individual grains as micro-intergrowths. It was also shown that the titanomagnetite (Ti-Mt) which amounts, in the Haruna rock, to over 90% of the total concentration of the ferromagnetic mineral contained, plays no appreciable role in producing the RTRM, for it usually coexists as separate grains. These facts are in accordance with the theoretical expectation that the magnetic interaction will be important only when the concentration of ferromagnetic minerals is abnormally higher than that found in common rocks [1], [11], [12]. The case in which different ferromagnetic minerals coexist with high concentration is realized more effectively and frequently when these minerals intergrow within an individual grain as in the Haruna-case than when the separate grains are gathered locally. The reason is the mean concentration of ferromagnetic minerals in rocks usually amounts to scarcely more than a few % and, moreover, the main ferromagnetic minerals concerned are the members of the ternary system $FeO$-$Fe_2O_3$-$TiO_2$ which is known to be very rich in phase transformations such as exsolutions.

Theoretical considerations of the nature of magnetic interaction have shown that, although it may act in a positive sense in some cases, it certainly acts negatively when the participating minerals coexist in parallel alternating planes as is found to result from the usual exsolution process in minerals [12]. Consequently, when such an exsolution takes place at a temperature higher than the Curie points of both the host and the exsoluted phases, the TRM acquired in subsequent weak-field cooling can be a RTRM. The most remarkable example of this case is the RTRM of Haruna, Sokota and Asio rocks and the artificially produced RTRM of heat treated minerals of Himesima iron sand [14].

Another example is the anomalous increase (AI) of TRM found in some natural Ti-Mt grains [15]. This phenomenon has recently been studied by Nagata and Ozima in detail [16]. When, on the other hand, the exsolution takes place at a temperature lower than the Curie point of any one or more of the phases concerned, the mechanism producing remanent magnetism would be somewhat different. Kawai, Kume, Sasajima and Yasukawa discovered some very interesting results on such cases by studying a large number of natural Ti-Mts [17]–[19]. We shall refer to these matters in **11** of this paper.

In the present work, a study was made of the relationship between TRM and the magnetic properties of various minerals of the ternary system $FeO$-$Fe_2O_3$-$TiO_2$ from the view point of the magnetic interaction among these minerals. Different combinations of various minerals of the ternary system were obtained by heat treating the natural ferromagnetic crystals of the Il-Ht series.

## 1. Preparatory Remarks

The ferromagnetism of the Il-Ht series ($x\,FeTiO_3 \cdot (1-x)Fe_2O_3$) was discovered rather recently [7], and the general magnetic and crystallographic characteristics have been studied by several authors on both natural and synthetic samples [20]-[22].* This series is ferromagnetic (or ferrimagnetic) when $0.75 > x > 0.55$, paramagnetic when $1 > x > 0.75$, and very weakly ferromagnetic (or parasitically ferromagnetic) when $0.55 > x > 0$, at ordinary temperatures. We will call these three groups ***ferromagnetic ilmenite***, ***ilmenite*** and ***titan-hematite*** respectively.

The occurrence of ferromagnetic ilmenite in nature is rather rare and even when it occurs its abundance in rocks is usually not more than one tenth of the coexisting Ti-Mt. According to Kuno [24], ferromagnetic ilmenite is found usually, as far as Japanese rocks are concerned, in hypersthene hornblende dacites. The reason for our adopting this series for the present study, in spite of its rather rare occurrence, is that it is precisely members of this series which produce the Haruna-type RTRM, and that the various significant combinations of minerals are more easily obtained from this series by heat treatments.

In this study, we have made full use of the knowledge on the fundamental properties of this series reported by Akimoto [21] and by Nagata and Akimoto [22]. Most of the following data in this section are from their papers. The properties of

*Table I Properties of the members of Ilmenite-Hematite (Il-Ht) series examined in the present study*

| Locality | Rock | $T_s$°C*₁ | Chemical Comp.*₂ | | | Crystal Parameters | | Curie Point | $I_s(T_0)$*₃ |
|---|---|---|---|---|---|---|---|---|---|
| | | | FeO | $Fe_2O_3$ | $TiO_2$ | $a_{rh}$ Å | $\alpha_{rh}$ | $T_c$°C | |
| Haruna | dacite pumice | 250~230 | 34.30 | 29.36 | 36.34 | 5.478 | 55°02′ | 260 | 23 |
| Sokota | iron sand | 300~$T_0$ | 38.00 | 27.79 | 34.21 | 5.483 | 55 01 | 250 | 26 |
| Himesima | horn. mica andesite | 300~$T_0$ | 35.53 | 20.70 | 43.77 | 5.489 | 54 56 | 190 | 29 |
| Gokurakuzi(1) | iron sand | 150~130 | 37.18 | 21.49 | 41.32 | 5.487 | 55 00 | 130 | 30 |
| ,, (2) | ,, | 70~ 50 | 38.47 | 17.48 | 44.05 | 5.494 | 54 57 | 70 | 24 |
| Asama | dacite pumice | 300~$T_0$ | 40.66 | 16.97 | 42.37 | 5.498 | 54 56 | 100 | 18 |
| Minakami | rhyolite tuff | 40~ 25 | 36.68 | 18.80 | 44.52 | 5.501 | 54 55 | 55 | 12 |
| Ore-ilmenite (Thailand) | ore minerals | — | 41.14 | 12.46 | 46.40 | 5.516 | 54 52 | 20 | 0.6 |

*₁ $T_s$: thermo-magnetic separation temperature.
*₂ in mol. %.
*₃ in emu/gr. at room temperature $T_0$.

the samples used in the present work are summarized in Table I. Just as in the case of Ti-Mt, the Curie point of ferromagnetic ilmenite contained in rocks is ranged over a certain temperature interval as illustrated by the thermo-magnetic separation spectra [7] of Fig. 1, a, b, c, d. Therefore the figures in Table I may be taken as concerned with the average of a whole sample: in most cases the average figures may be considered

* For instance, Pouillard made some pioneering research on this series, but not reported about the ferromagnetic nature [23].

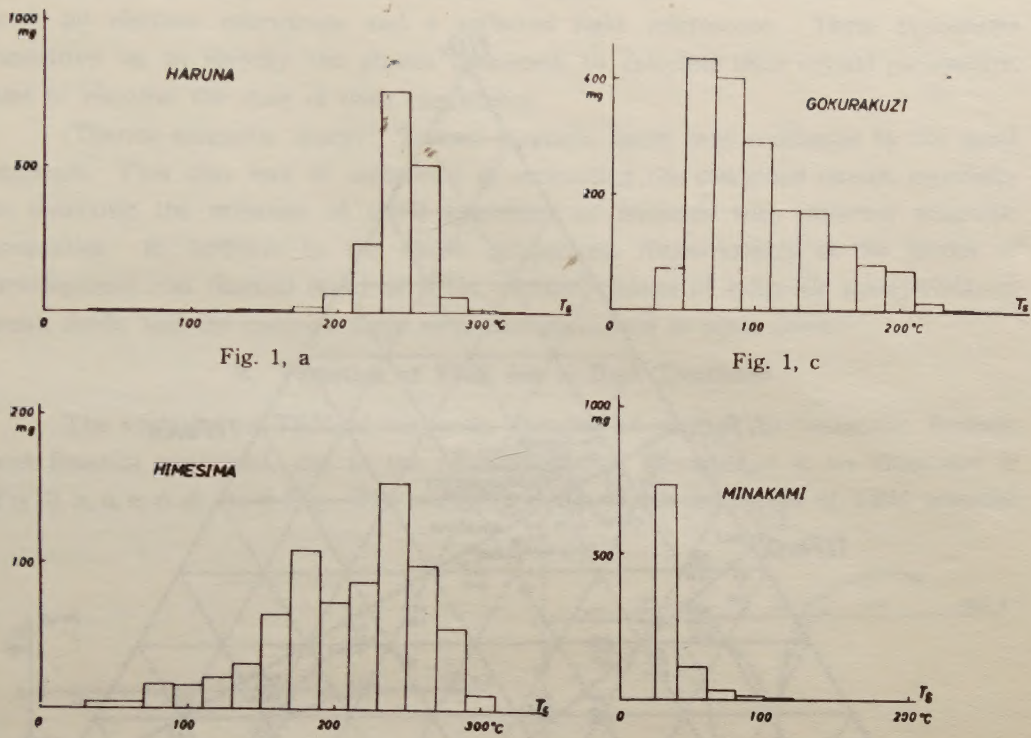



Fig. 1, a

Fig. 1, c

Fig. 1, b

Fig. 1, d

Fig. 1 Thermo-magnetic separation spectra of ferromagnetic ilmenites of Haruna, Himesima, Gokurakuzi and Minakami samples (after Nagata and Akimoto).

as those of the principal component of thermo-magnetic separation spectra. Fig. 2 is the plot of the chemical composition of the used samples on the $FeO$-$Fe_2O_3$-$TiO_2$ ternary diagram. (For reference, all the thermo-magnetically separated components are plotted for the Gokurakuzi sample.) In Fig. 2, the empirical relations [21], [22] between the chemical composition and the Curie point ($T_c$) and the lattice parameter ($a_{rh}$) and ($a_c$) of the series Il-Ht and magnetite-ulvöspinel are also shown: from these relations one may obtain the chemical composition, $T_c$ and $a_{rh}$ or $a_c$ of a sample of these series, if one knows any one of the three variables.

## 2. Experimental Procedures

(Heat treatments): The samples, packed in an open-air quartz tube with an asbestos plug or in a sealed quartz tube evacuated to as low as $10^{-3}$ mmHg., were inserted into an electric furnace after the temperature in the furnace had setteled at the designated temperature, and, after having been kept there for a designated length of time, taken out and cooled in air. The length of the heat treatment was 10 minutes in most cases. In the present paper, the following notations will be used: for instance, *Haruna* (250°C$>T_s>$230°C, 850°C, AIR or VAC, 10 min.). This is to identify the Haruna sample, having the thermo-magnetic separation temperature $T_s$ between 250°C and 230°C, which was heat-treated in the open-air or evacuated sealed tube at 850°C for 10 minutes.

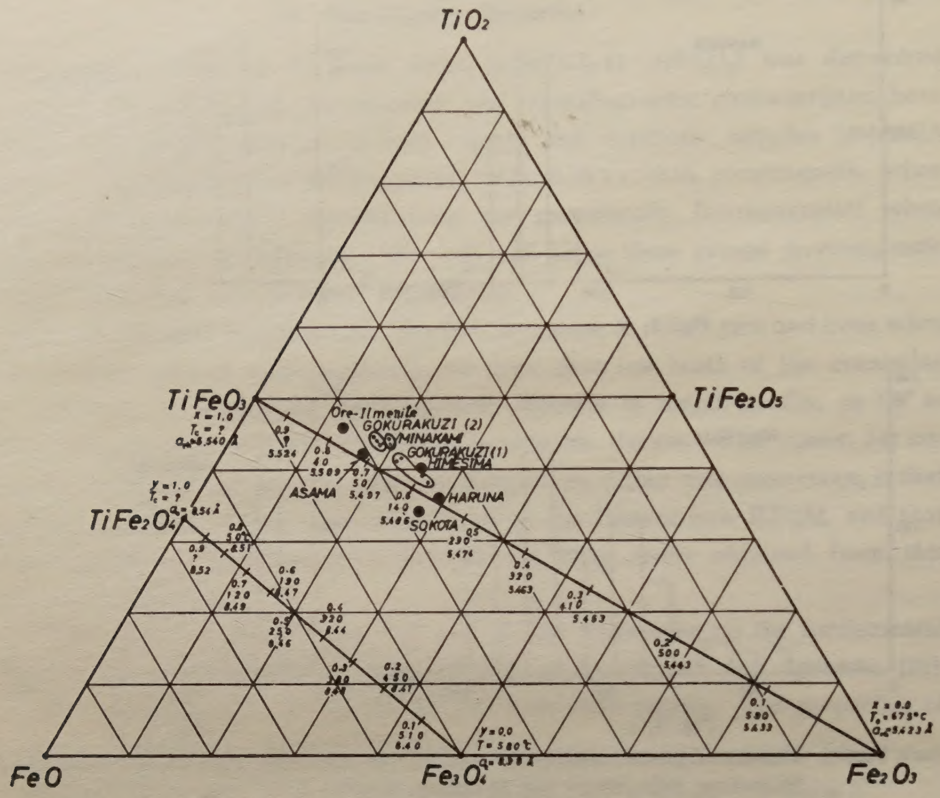



Fig. 2 Plot of the chemical composition of the ferromagnetic ilmenites examined in the present study, and the relations between chemical composition and Curie point ($T_c$) and lattice parameters ($a_{rh}$ and $a_c$) of the joins ilmenite-hematite and magnetite-ulvöspinel established by Nagata and Akimoto.

(TRM): The thermo-remanent magnetism was produced, as a rule, when each heat-treated sample cooled from the treatment temperature by putting the axis of the tube parallel to the geomagnetic field in the laboratory. TRM was measured at room temperature ($T_o$) by means of an astatic magnetometer.

(Chemical analysis): Since it was anticipated that, during the heat treatment, not only phase transformations but also chemical changes such as oxidation and reduction would take place, chemical analysis of the treated samples was carried out whenever necessary. All the chemical work has been carried out by Mr. Katsura at the Tokyo Institute of Technology. Since the duration of the treatment may be too short for the sample to attain true equilibrium, particularly where chemical reactions were concerned, we by no means claim that the resultant state brought about by the heat treatment in the present work represents the true equilibrium state at the treatment temperature.

(X-ray analysis and microscopic observation): As the chemical analysis determines only the total composition of the sample, we also conducted X-ray analysis of the samples by Norelco spectrometer and made observations of the polished surfaces

with an electron microscope and a reflected light microscope. These procedures permitted us to identify the phases contained, to calculate their crystal parameters, and to visualize the state of their coexistence.

(Thermo-magnetic study): Thermo-magnetic study was conducted by the usual methods. This also was of assistance in estimating the contained phases, especially in detecting the presence of small quantities of minerals with different magnetic properties. In addition to the above procedures, measurements of the modes of development and thermal decay of TRM, thermal change of magnetic susceptibility in weak fields, and the coercive force were also conducted in some cases.

## 3. Variation of TRM due to Heat Treatment

The variation of TRM of the seven samples of natural ferromagnetic ilmenite and ilmenite examined, due to the heat treatments described in **2**, are illustrated in Fig. 3, a, b, c, d, $d_1$, $d_2$, e, f, g. The ordinates represent the intensities of TRM acquired

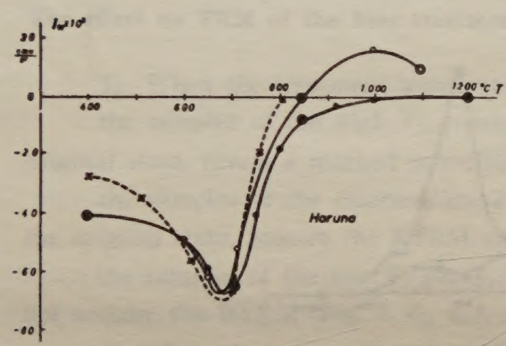



**Fig. 3, a Variation of TRM of the Haruna sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles).**

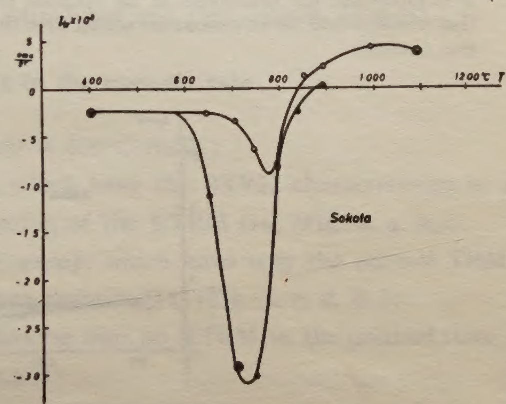



**Fig. 3, b Variation of TRM of the Sokota sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles).**

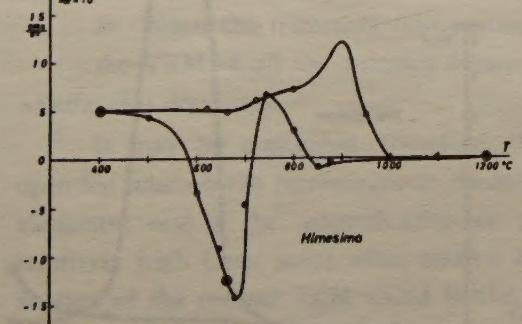



**Fig. 3, c Variation of TRM of the Himesima sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles).**

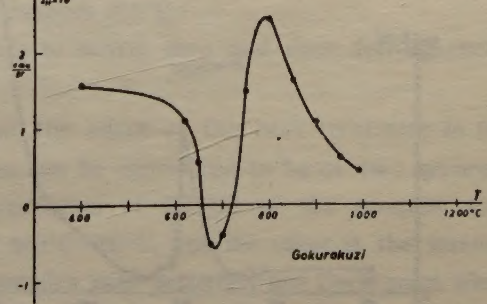



**Fig. 3, d Variation of TRM of the Gokurakuzi sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube.**

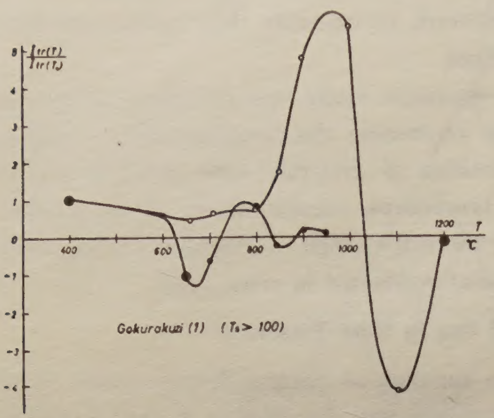



Fig. 3, $d_1$ Variation of TRM of the Gokurakuzi (1) ($T_s$>100°C) sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles).

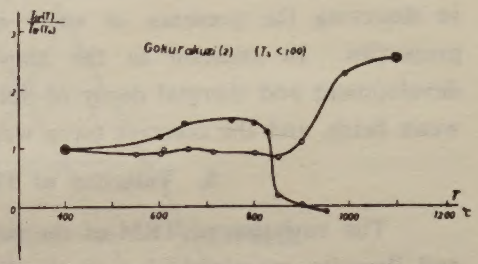



Fig. 3, $d_2$ Variation of TRM of the Gokurakuzi (2) ($T_s$<100°C) sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles).

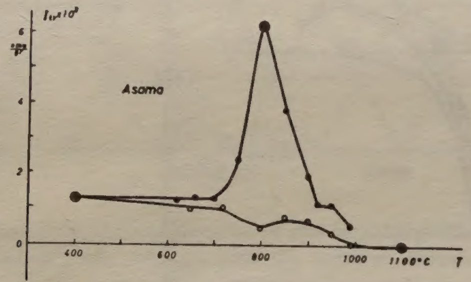



Fig. 3, e Variation of TRM of the Asama sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles).

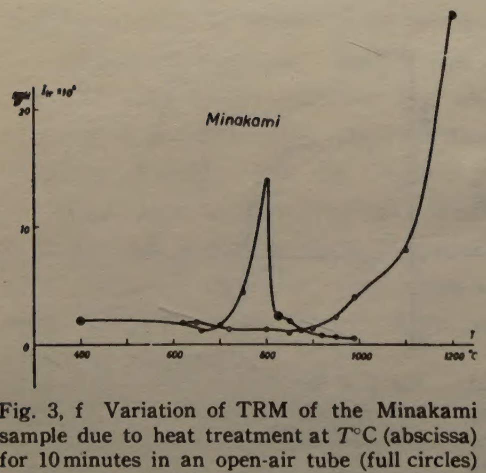



Fig. 3, f Variation of TRM of the Minakami sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles).

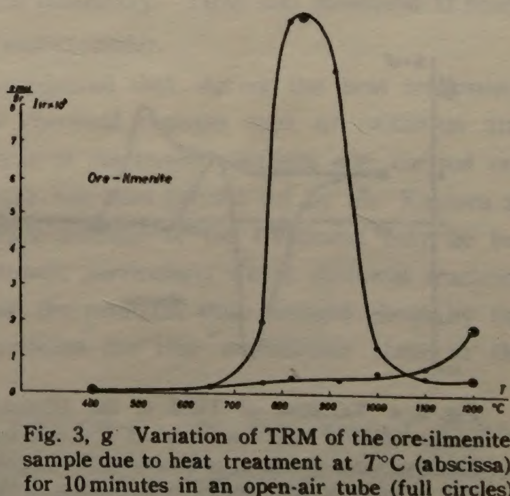



Fig. 3, g Variation of TRM of the ore-ilmenite sample due to heat treatment at $T$°C (abscissa) for 10 minutes in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles).

in the geomagnetic field after being heat treated at various temperatures shown on the abscissa. The full circles and the hollow circles correspond to the heat treatment in the open-air tube and the evacuated sealed tube respectively. The double circles stand for the samples on which at least one of the chemical, crystallographic, thermomagnetic or microscopic examinations was conducted. In Fig. 3, a, the cross marks show the final value of TRM attained after repeated heat treatments in the open-air tube at respective temperatures.

Although the curves in these figures seem to be rather complicated, we may be able to summarize the principal features of them as follows: To do this, it will be convenient to classify the examined samples into the following three groups according to their respective Curie points ($T_c$), (*cf.* Table I)

***high $T_c$ group:*** Haruna and Sokota samples

***intermediate $T_c$ group:*** Himesima and Gokurakuzi (1) samples

***low $T_c$ group:*** Gokurakuzi (2), Asama, Minakami and Ore-ilmenite samples.

**The effect on TRM of the heat treatment in the open-air tube.**

1) When the treatment temperature is 600°C–700°C;

the samples of the ***high $T_c$ group***, which have the RTRM characteristics in the original state, reveal a marked intensification of the RTRM [14] (Fig. 3, a, b.);

the samples of the ***intermediate $T_c$ group***, which have only the normal TRM in the original state, acquire the RTRM characteristics [14] (Fig. 3, c, d, $d_1$.);

the samples of the ***low $T_c$ group***, having also no RTRM in the original state, do not acquire the RTRM (Fig. 3, $d_2$, e, f, g.).

2) When the treatment temperature is 750°C–850°C;

the samples of the ***high $T_c$ group***, show a monotonous decrease in the RTRM;

the samples of the ***intermediate $T_c$ group*** which acquired the RTRM in 1), again show the normal TRM;

the samples of the ***low $T_c$ group*** reveal a very remarkable intensification in the normal TRM.

3) When the treatment temperature exceeds 850°C;

the TRM of all the samples diminishes to nearly zero and their ferromagnetic nature also decreases.

It may be concluded, therefore, that the effect of the heat treatment in the open-air tube on the ferromagnetic ilmenites can be considered to be of two separate elements: one is the intensification or production of RTRM of the samples with relatively high Curie point when treated at 600°C–700°C, and the other is the intensification of the normal TRM found in the samples with relatively low Curie point when the treatment temperature is about 750°C–850°C. Hence, the samples of the ***intermediate $T_c$ group*** having widespread thermo-magnetic separation spectra (Fig. 1, b, c.) can be considered as being affected by both of the two elements mentioned above. The results shown in Fig. 3, d, $d_1$, $d_2$ may be considered as the evidence of this point of view.

**The effect on TRM of the heat treatment in the evacuated sealed tube.**

1) When the treatment temperature is 600°C–700°C;

the samples of the *high $T_c$ group* reveal an intensification of RTRM, but the intensification of RTRM of the Sokota sample is conspicuously less than when treated in the open-air tube;

the samples of the *intermediate $T_c$ group* do not acquire RTRM characteristics, being in sharp contrast to when treated in the open-air tube;

the samples of the *low $T_c$ group* show no appreciable change in TRM.

2) When the treatment temperature exceeds 700°C, the features are rather complicated;

the samples of the *high $T_c$ group* show an increase in the normal TRM, but when the treatment temperature is raised more the normal TRM begins to decrease;

the samples of the *intermediate $T_c$ group* show the same features as the *high $T_c$ group*. But, the *Gokurakuzi* (1) ($T_s$>100°C, 1100°C, VAC, 10 min) sample abruptly acquires a RTRM. This is a new type of RTRM, having a somewhat different mechanism from that of the Haruna-type RTRM: the magnetic interaction is between the ferromagnetic ilmenite and the Ti-Mt produced by the reduction of the original sample, as will be shown later.

The samples of the *low $T_c$ group* show a steady increase in the normal TRM, the Asama sample being an exception.

The effect of heat treatment in the sealed evacuated tube, therefore, seems to be less systematically related to the Curie point of the sample than in the case of the treatment in the open-air tube. In the following discussions, we will try to show how these features may be explained in terms of magnetic interaction.

## 4. Results of Chemical, Crystallographic and Thermo-magnetic Analyses, and Microscopic Examination.

To interpret the various phenomena described above, the chemical, crystallographic, and thermo-magnetic analyses and microscopic examination were made on some of the samples: namely the samples represented by the double circles in Fig. 3, a, b, c, d, $d_1$, $d_2$, e, f, g. The results are summarized in Fig. 4, a, b, c, d, e and in Table II. These data will be frequently referred to in the later sections. In Fig. 4, the dotted lines show the phases identified by X-ray analysis. These dotted lines have been plotted, as mentioned already, by making use of the empirical relations between the lattice parameters and the chemical composition for the Ti–Mt and Il–Ht series shown in Fig. 2. By doing this their Curie points are estimated simultaneously. The Curie points thus estimated were actually found to be in fairly good agreement with those obtained directly by thermo-magnetic study and listed in Table II. As for rutile and pseudobrookite, we neglected the solid solutions.

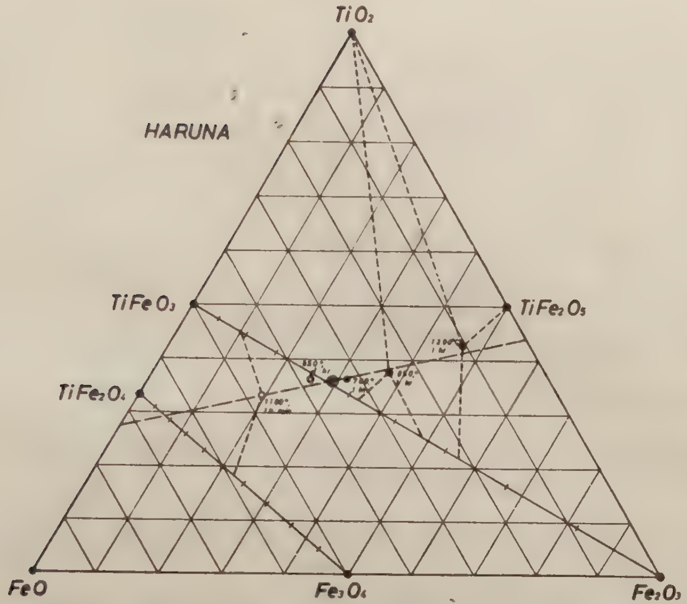



Fig. 4, a Results of chemical and crystallographic analyses of the Haruna sample heat treated in an open-air tube (full circles) and in an evacuated sealed tube (hollow circles). The dotted lines and the broken line show the chemical composition of coexisting phases estimated from X-ray data and the theoretical reduction-oxidation line, respectively. The double circle represents the original sample.

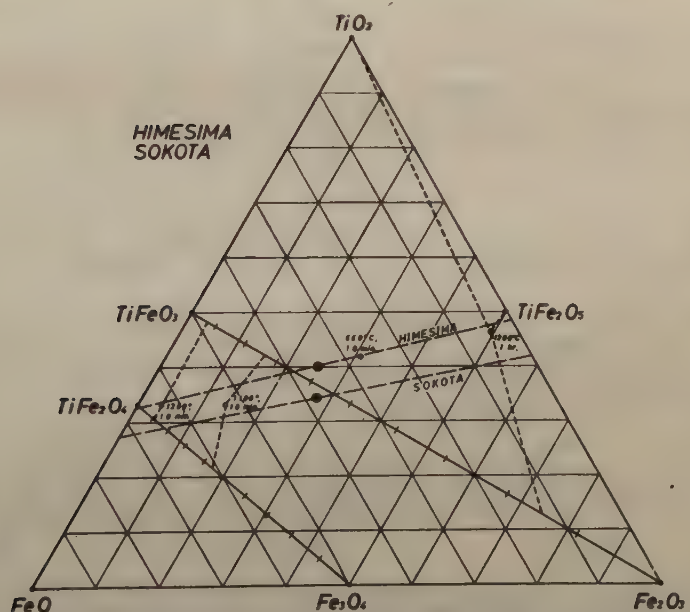



Fig. 4. b Results of chemical and crystallographic analyses of the Sokota and Himesima samples heat treated in open-air (full circles) and in an evacuated sealed tube (hollow circles). The dotted lines and the broken lines show the chemical composition of coexisting phases estimated from X-ray data and the theoretical reduction-oxidation lines, respectively. The double circles represent the original samples.

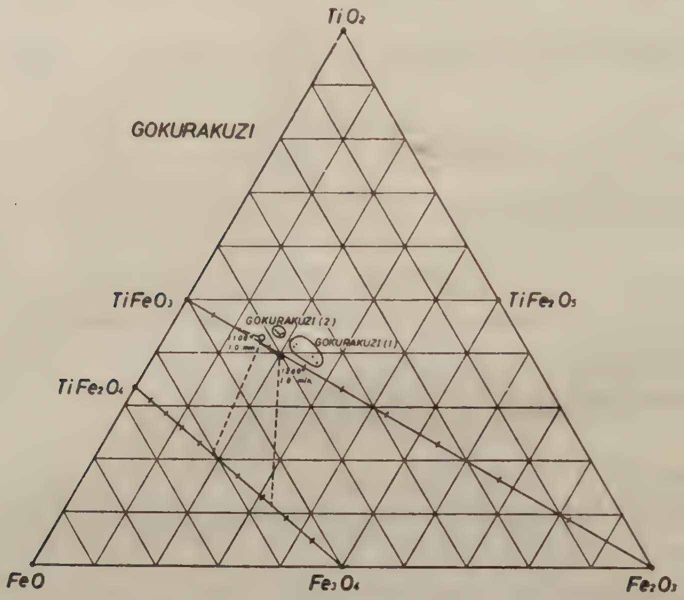



Fig. 4, c Results of chemical and crystallographic analyses of the Gokurakuzi (1) and Gokurakuzi (2) samples heat treated in an evacuated sealed tube. The full circlets represent the thermo-magnetically separated original sample.

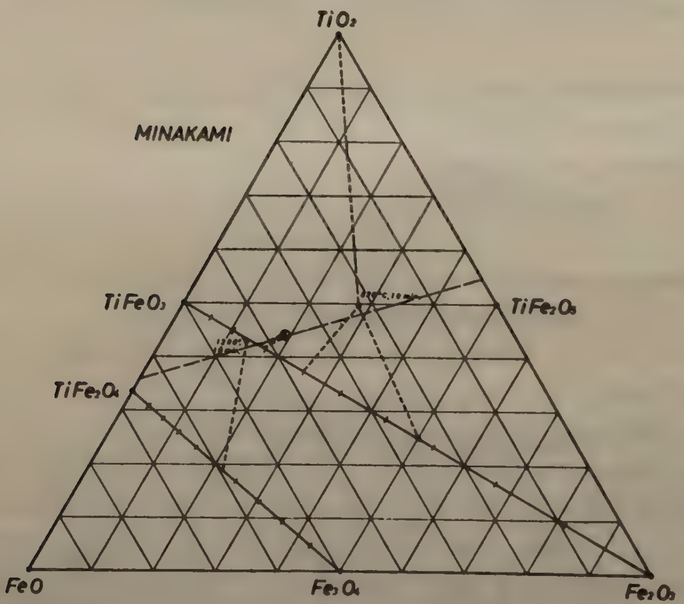



Fig. 4, d Results of chemical and crystallographic analyses of the Minakami sample, heat treated in an open-air tube (full circle) and in an evacuated sealed tube (hollow circle). The dotted lines and the broken line show the chemical composition of coexisting phases estimated from X-ray data and the theoretical reduction-oxidation line, respectively. Two full circlets in a circle represent the thermo-magnetically separated original sample.

*Table* II

| Locality | $T_s$°C*1 | Heat Treatment | Chemical Comp.*2 FeO | $Fe_2O_3$ | $TiO_2$ |
|---|---|---|---|---|---|
| Haruna | 250~230 | original | 31.8 | 27.1 | 33.8 |
| | ,, | 700°, AIR, 1 hr. | 32.1 | 31.8 | 36.1 |
| | ,, | 850 , AIR, 1 hr. | 25.4 | 36.6 | 38.0 |
| | ,, | 1200 , AIR, 1 hr. | 10.3 | 47.1 | 42.6 |
| | ,, | 700 , VAC, 1 hr. | 34.7 | 28.9 | 36.4 |
| | ,, | 850 , VAC, 1 hr. | 38.2 | 25.5 | 36.3 |
| | 230~210 | 1100 , VAC, 1 hr. | 37.5 | 19.4 | 33.1 |
| Sokota | 350~ $T_o$ | original | 38.0 | 27.8 | 34.2 |
| | ,, | 1100 , VAC, 10 min. | 53.1 | 13.8 | 33.1 |
| Himesima | 250~230 | original | 34.7 | 25.5 | 39.8 |
| | ,, | 660 , AIR, 10 min. | 27.3 | 31.0 | 41.7 |
| | ,, | 1200 , AIR, 1 hr. | 4.2 | 49.5 | 46.3 |
| | 210~170 | 1200 , VAC, 10 min. | 63.9 | 3.8 | 32.3 |
| Gokurakuzi (1) | 150~130 | original | 37.2 | 21.5 | 41.3 |
| | >100 | 1200 , VAC, 10 min. | 41.2 | 20.0 | 39.8 |
| ,, (2) | 70~ 50 | original | 38.5 | 17.5 | 44.1 |
| | 100~ $T_o$ | 1100 , VAC, 10 min. | 41.4 | 15.5 | 43.1 |
| Asama | 300~ $T_o$ | original | 40.6 | 17.0 | 42.4 |
| | ,, | 1100 , VAC, 10 min. | 59.4 | 4.1 | 36.5 |
| Minakami | 40~ 25 | original | 36.7 | 18.8 | 44.5 |
| | ,, | 820 , AIR, 10 min. | 22.3 | 28.4 | 49.3 |
| | 100~ $T_o$ | 1200 , VAC, 10 min. | 43.5 | 13.2 | 43.3 |
| Ore-ilmenite (Thailand) | | original | 41.1 | 12.5 | 46.4 |
| | | 850 , AIR, 10 min. | 31.1 | 20.3 | 48.6 |
| | | 1200 , AIR, 10 min. | 18.6 | 29.5 | 51.9 |
| | | 1200 , VAC, 10 min. | 44.1 | 11.1 | 44.8 |

*1 Thermo-magnetic separation temperature before heat treatment. $T_o$ is the room temperature.
*2 Mol. %
*3 Result obtained by X-ray analysis and microscopic examination; 2 ferro-imenites means the
*4 $T_o >$ means that $T_o > T_\varphi$.

| Coexisting*3 Minerals | Il-Ht series | | | Ti-Mt series. | | $I_s(T_0)$ $\frac{emu}{gr}$ | TRM |
|---|---|---|---|---|---|---|---|
| | $a_{rh}$ Å | $\alpha_{rh}$, | $T_c$ °C | $a_r$ Å | $T_c$ C | | |
| 2 ferro-ilmenites | 5.478 | 55 02 | 250 | | | 23 | reverse |
| ,, | 5.480 | 55 01 | 210 | | | 27 | reverse |
| 2 ferro-ilmenites | 5.473 | 55 06 | 210 | | | 4 | reverse |
| titan-hematite | 5.459 | 55 08 | 450 | | | | |
| rutile | | | | | | <1 | 0 |
| titan-hematite | 5.451 | 55 07 | 420 | | | | |
| pseudobrookite, rutile | | | | | | | |
| 2 ferro-ilmenites | 5.479 | 55 02 | 185 | | | 23 | reverse |
| 2 ferro-ilmenites | 5.483 | 55 02 | 160 | | | 2 | normal |
| Ti-Mt (faint) | | | | | 400 | | |
| ilmenite | 5.512 | 54 55 | $T_0>$*4 | | | 25 | normal |
| Ti-Mt | | | | 8.451 | 240 | | |
| 2 ferro-ilmenites | 5.483 | 55 02 | 250 | | | 26 | reverse |
| ilmenite | 5.500 | 55 27 | $T_0>$ | | | 14 | normal |
| Ti-Mt | | | | 8.467 | 150 | | |
| 2 ferro-ilmenites | 5.480 | 55 02 | 250 | | | 30 | normal |
| ,, | | | | | | 29 | reverse |
| pseudobrookite | | | | | | | |
| titan-hematite | 5.438 | 55 14 | | | | 0 | 0 |
| rutile (faint) | | | | | | | |
| ulvöspinel | | | | 8.521 | $T_0>$ | 0 | 0 |
| ilmenite | 5.530 | 54 48 | $T_0>$ | | | | |
| 2 ferro-ilmenites | 5.487 | 55 00 | 130 | | | 30 | normal |
| ferro-ilmenite | 5.499 | 54 59 | 90 | | | 34 | reverse |
| Ti-Mt | | | | 8.420 | 450 | | |
| 2 ferro-ilmenites | 5.494 | 54 57 | 70 | | | 24 | normal |
| ilmenite | 5.512 | 54 54 | $T_0>$ | | | 14 | normal |
| Ti-Mt | | | | 8.462 | 280 | | |
| 2 ferro-ilmenites | 5.498 | 54 56 | 100 | | | 18 | normal |
| ulvöspinel | | | | 8.514 | $T_0>$ | 0 | normal |
| ilmenite | 5.524 | 54 53 | $T_0>$ | | | | |
| 2 ferro-ilmenites | 5.501 | 54 55 | 55 | | | 12 | normal |
| ferro-ilmenite | 5.486 | 55 07 | 160 | | | | |
| titan-hematite | 5.456 | 55 12 | | | | 8 | normal |
| rutile | | | | | | | |
| ilmenite | 5.512 | 54 52 | $T_0>$ | | | 4 | normal |
| Ti-Mt | | | | 8.454 | 200 | | |
| ilmenite | 5.516 | 54 52 | $T_0>$ | | | <1 | 0 |
| ferro-ilmenite | 5.492 | 55 02 | 100 | | | | |
| titan-hematite | 5.468 | 55 06 | 150 | | | 5 | normal |
| rutile | | | | | | | |
| ferro-ilmenite | 5.483 | 55 17 | 80 | | | | |
| titan-hematite | 5.437 | 55 13 | 500 | | | 3 | normal |
| rutile | | | | | | | |
| pseudobrookite | | | | | | | |
| ilmenite | 5.511 | 54 58 | $T_0>$ | | | 3 | normal |
| Ti-Mt (faint) | | | | | 140 | | |

**Haruna-type exsolution of two ferromagnetic ilmenites.**

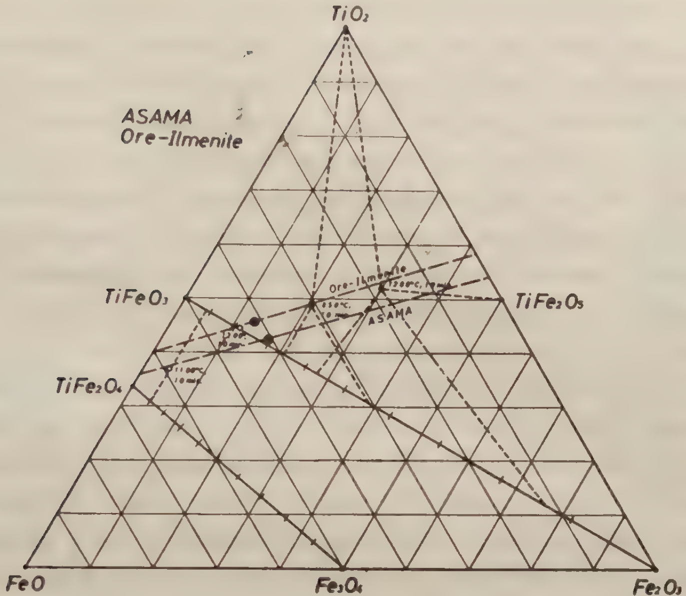



Fig. 4, e Results of chemical and crystallographic analyses of the Asama and Ore-ilmenite samples heat treated in an open-air tube (full circles) and in an evacuated sealed tube (hollow circle). The dotted lines and the broken lines show the chemical composition of coexisting phases estimated from X-ray data and the theoretical reduction-oxidation lines. The double circles represent the original samples.

## 5. Interpretation of Intensification and Artificial Production of RTRM by Heat Treatment at 600°C–700°C in the Open-air Tube.

As seen in Fig. 3, a, b, c, d, $d_1$, the RTRM of the Haruna and Sokota samples is remarkably increased while the Himesima and Gokurakuzi (1) samples acquire the Haruna-type RTRM, through the heat treatment in the open-air tube at 600°C–700°C for 10 minutes [14]. Naturally, the problem to be dealt with in this section is closely related to that of the Haruna-type RTRM itself. The latter problem was studied in the previous paper in considerable detail [12], the main results of which will be summarized here as follows: This type of RTRM is considered to be originated from the magnetic interaction between two ferromagnetic Il-Ht solid solutions closely intergrown within each grain as observed by electron microscope, and the properties of these solid solutions, such as lattice parameters and Curie points should be very similar. For instance, their Curie points in the case of Haruna sample would be about 320°C–250°C and 250°C–200°C or lower. In the previous paper, the nature of these two constituents were not determined any further.

Judging from the fact that the ferromagnetic ilmenite of the Haruna sample contains more $Fe_2O_3$ than the limiting member of solubility of hematite into ilmenite $\left(\frac{1}{3}Fe_2O_3 \cdot \frac{2}{3}TiFeO_3\ [20], [23]\right)$, one may readily expect that the component with the higher Curie point (the $A'$-component in [12]) would be a member of titan-hematite. But this is not true, because titan-hematite is not ferromagnetic

in the ordinary sense [20]~[22], and, even if it is, the Curie point would be too high (at least 400°C) to meet the observed Curie point of the $A'$-component of Haruna sample. Therefore, both of these ferromagnetic minerals should belong to the ilmenite-side of the solvus curve of the system Il-Ht. An intergrowth, however, of components belonging to the same side of a solvus curve is impossible as far as the equilibrium state is concerned. In actuality, the fact that the nature of the RTRM of the samples examined here is extremely sensitive to the heat treatment of relatively short duration indicates that we are dealing with non-equilibrium or metastable states of the minerals concerned. In the following discussion, therefore, we will base ourselves on this hypothesis.

In Fig. 4, a, are shown the results of chemical analysis of the Haruna sample treated at various temperatures for *1 hour*. One can see that the oxidation by the heat treatment at 700°C in the open-air tube is very little. Since the duration of the treatments in most of other cases in the present study is only ***10 minutes***, we may assume that the oxidation due to the heat treatments of other samples will generally be not more radical than what is shown in Fig. 4, a. As a result, the oxidation taking place during the heat treatments concerned in this section, namely 600°C–700°C for 10 minutes, would also be small for the other samples.* Same will be said for the reduction taken place during the heat treatment at 600°C–700°C in the evacuated sealed tube. The effect of such slight oxidation and reduction was found to be important in the cases of the Sokota, Himesima and Gokurakuzi (1) ($T_s$>100°C) samples, because the curves corresponding to the treatments in the open-air tube and the evacuated sealed tube in Fig. 3, b, c, $d_1$ show very different features at 600°C–700°C. We will discuss this point later in this section.

Another important experimental result accompanying the heat treatment at about 600°C–700°C is illustrated in Fig. 5: the Curie point of the Haruna ferromagnetic ilmenite decreases after the heat treatment at 700°C in both the open-air tube and the evacuated sealed tube. Since it has already been proved that little oxidation has taken place during the treatment in the open-air tube, it seems to be an evident contradiction of the empirical relation which states that the Curie point becomes higher when the hematite content increases in the Il-Ht series, as shown in Fig. 2. Such decrease in Curie point was observed in other samples of the ***high $T_c$ and intermediate $T_c$ groups***, as will be seen in Fig. 9, a.

Taking the various data described above into account, we will present a model, though still hypothetical, which explains the intensification and the artificial production of RTRM of the samples of the *high and intermediate $T_c$ groups*, and the non-produc-

* According to Curnow and Parry [25] and Ishikawa and Sawada [26], the oxidation of ilmenites is fairly radical at 600°C in the open air. The reason why oxidation in our treatments is small is because the samples in our case are packed in a quartz tube with an asbestos plug. Actually, when the treatment was performed in free-circulating open air, the oxidation was found to be more radical. Such an example is the *Himesima* (250°C>$T_s$>230°C, 660°C, AIR, 10 min.) sample shown in Fig. 4, b.

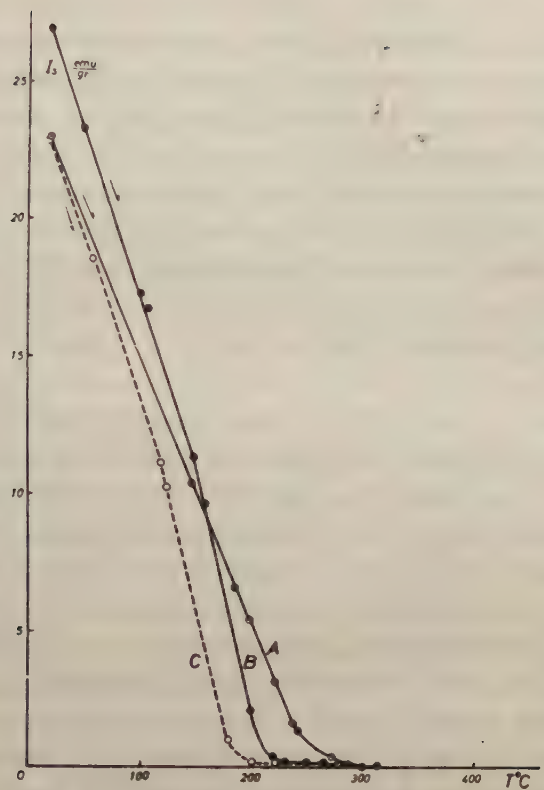



Fig. 5 $I_s$ $(T)$ curves of Haruna sample:
A): *Haruna* (250°C$>T_s>$230°C, original)
B): *Haruna* (250°C$>T_s>$230°C, 700°C, AIR, 1 hr)
C): *Haruna* (250°C$>T_s>$230°C, 700°C, VAC, 1 hr)
Curie point decreases after the heat treatment.

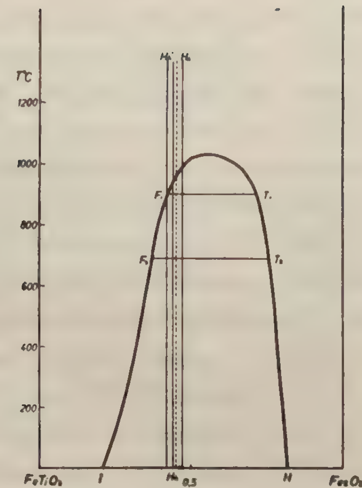



Fig. 6 Probable form of phase relationship in system ilmenite-hematite. The dotted line is the boundary separating ferromagnetic region (left) and parasitic ferromagnetic region (right), after Nagata and Akimoto.
$H_A$: *Haruna* (original)
$H'_A$: *Haruna* (600°C-700°C, AIR)
$H''_A$: *Haruna* (600°C-700°C, VAC)

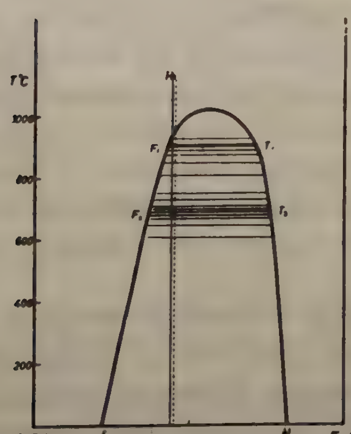



Fig. 7 Illustration of coexisting phases in Haruna sample originated from the "partial exsolution."

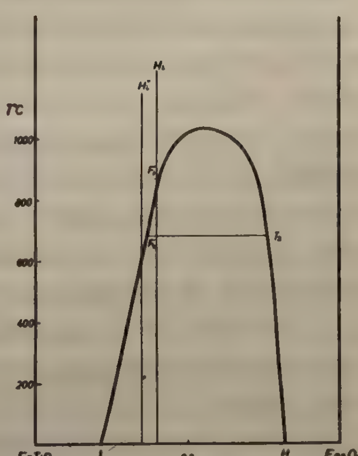



Fig. 8 Probable form of phase relationship in system ilmeinte-hematite:
$H_i$: *Himesima* (original)
$H''_i$: *Himesima* (600°C-700°C, VAC)

tion of RTRM in the samples of the *low $T_c$ group*. Fig. 6 is the probable phase relationship of the Il-Ht system proposed by Nicholls [27] and modified by the present author to meet our experimental results. In this diagram, $H_A$ represents the composition of the Haruna sample in its original state. According to Kuno [28], the mother rocks of the ferromagnetic ilmenites examined here are volcanic in origin and their crystallization temperature should range between 800°C–900°C approximately. We assume this temperature to be about 900°C in this paper tentatively. Therefore, the mineral at the crystallization temperature would have exsolved into $F_1$ and $T_1$, their relative contents being $H_AT_1 : H_AF_1$ where $H_AT_1 \gg H_AA_1$. During the subsequent cooling process, the exsolution should have proceeded along $F_1I$ and $T_1H$. But, as the cooling rate of such volcanic rocks is estimated to be very high, the exsolution process may not have sufficient time to attain the true equilibrium state at each temperature. In other words, the exsolution at each temperature level may have always been incomplete. If such a " partial exsolution " has actually taken place, the Haruna sample as we see at present should contain, in each grain, many ferromagnetic ilmenites with nearly continuous chemical composition. However, it is highly probable that in the process of total cooling, there was a temperature level where the exsolution was most effective. If such was the case, the components of the Haruna sample in the present state may be, aside from the small amount of titan-hematites ($T_1$ and $T_2$), roughly classified into two parts, $F_1$ and $F_2$, which would be closely intergrown with each other. Fig. 7 shows the probable model of the Haruna sample deduced schematically from the above hypothetical processes. We regard these two classes of ferromagnetic ilmenites as the major ferromagnetic minerals participating in the production of RTRM of the Haruna-type. We will further show that the above model affords a satisfactory explanation for the various experimental results. Now, let us assume that the temperature at which the most effective exsolution took place was 680°C–700°C. Then, when the sample undergoes the heat treatment at these temperatures, the $F_2$ component should develop at the expense of the $F_1$ component. If, further, the proportion of $F_1$ and $F_2$ after the heat treatment is nearer to the optimum proportion for producing RTRM than in the original one, the intensification of RTRM can be explained. The slight oxidation and reduction taking place during the heat treatments will displace the total composition of the sample from $H_A$ to $H_A'$ and $H_A''$ respectively (Fig. 6). But, as may readily be shown, such slight changes in the total composition do not alter the principal effects of the heat treatment in the Haruna case. The observed lowering of the Curie point due to the treatment, even in the open-air tube, will also be explained quite readily.

Now, let us extend this hypothetical model to the cases of the samples of the *intermediate $T_c$ group*, such as the Himesima and Gokurakuzi (1) ($T_s > 100$°C) samples. They, having no RTRM in the original state, acquire RTRM characteristics during the heat treatment at 600°C–700°C in the open-air tube, but not in the evacuated sealed tube (Fig. 3, c, d, $d_1$). In Fig. 8, $H_t$ represents the original average composition of the ferromagnetic ilmenite of the Himesima sample. As in the case of the Haruna sample,

we assume tentatively that the crystallization temprature of the mother rock of Himesima sample is about 900 C and that the subsequent rapid cooling resulted in the same "partial exsolution" as explained above. The well developed exsolution lamellae shown in the previous paper may be due to this partial exsolution. As the original Himesima sample shows normally directed TRM only, the magnetic interaction in this sample should not be strong enough to produce RTRM. However, when the sample is treated in the open-air tube at temperatures as high as 600°C–700°C, the exsolution of $F_2$ rapidly proceeds at the expense of $F_1$ and this process may result in the production of RTRM in the same manner as in the intensification of RTRM of the Haruna sample. The results of an experiment conducted on the Himesima (250°C>$T_s$>230°C) sample are summarized in Fig. 9, a, b: in Fig. 9, b abscissa denotes the temperature of heat treatment. In this experiment, the heat treatment was conducted in a free-circulating open-air condition. Therefore, the degree of oxidation was more radical than in the cases of the treatment in the tube with an asbestos plug (see Fig. 4, b). The lowering of the Curie point, irrespective of the oxidation, and the increase in the coercive force $H_c(T_0)$ and decrease in the susceptibility $\chi(T_0)$ due to the heat treatments are clearly seen. In the case where the heat treatment was conducted in the evacuated sealed tube, the reduction would have displaced the line representing the total composition of the sample to $H_i''$ in Fig. 8. Therefore, in this case, the heat treatment at 600°C–700°C will not promote the exsolution process but homogenization instead which would disfavour the occurrence of RTRM.

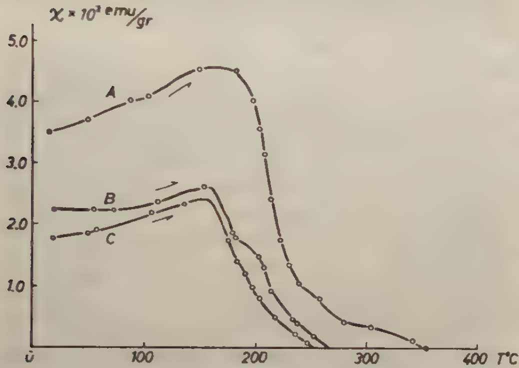



Fig. 9, a Thermal change of magnetic susceptibility of Himesima sample in $H$=7.0 Oe.
A): *Himesima* (250°C>$T_s$>230°C, original)
B): *Himesima* (250°C>$T_s$>230°C, 600°C, AIR, 10 min.)
C): *Himesima* (250°C>$T_s$>230°C, 660°C, AIR, 10 min.) showing RTRM
Curie point decreases by the heat treatment.

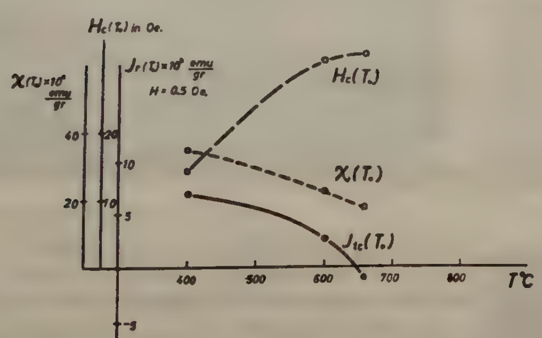



Fig. 9, b Variations of $H_c(T_0)$, $\chi(T_0)$ and $Jtr$ ($T_0$) due to the heat treatment at $T$°C (abscissa) in air for 10 minutes.
Sample: *Himesima* (250°C>$T_s$>230°C)
$H_c$ ($T_0$): coercive force at room temperature ($T_0$)
$\chi(T_0)$: magnetic susceptibility at $T_0$ in $H$=7.0Oe.
$Jtr$ ($T_0$): TRM at $T_0$, produced in $H$=0.5 Oe.

Similar expalnations will readily be applied to the Sokota and Gokurakuzi (1) ($T_s$>100°C) samples.

Finally, the non-production of

RTRM in the samples of the *low* $T_c$ *group* (Minakami, Gokurakuzi (2), Asama, and Ore-ilmenite samples) subjected to the same heat treatment will be explained (see Fig. 3, $d_2$, e, f, g.). The original composition of these samples are indicated by $M$, $G$, $A$, and $I$ in Fig. 10. The mineral composition in the original state will be, in the case of the Minakami sample for example, represented by $M$, $F_1$ and $T_1$, provided that the "partial exsolution" would have taken place at a temperature as low as, say, 400°C. Since these samples show no RTRM in their original state, the magnetic interaction between $M$ and $F_1$ must be too weak to produce any RTRM. It would be, then, easily understood that the treatment at 600°C–700°C would not promote the separation of $M$ and $F_1$, even when the oxidatien due to the treatment in the open-air tube would displace the composition to $M'$.

Plates 1, a, b, c, and 2, a, b, c show examples of the exsolution lamellae, observed through the electron and reflected light microscopes. The well

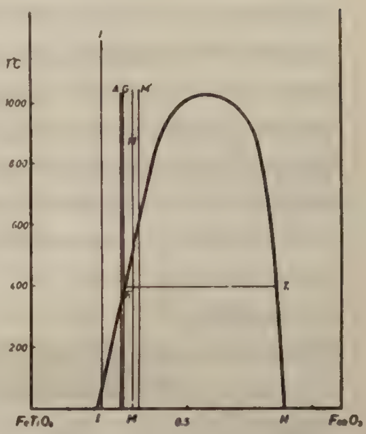
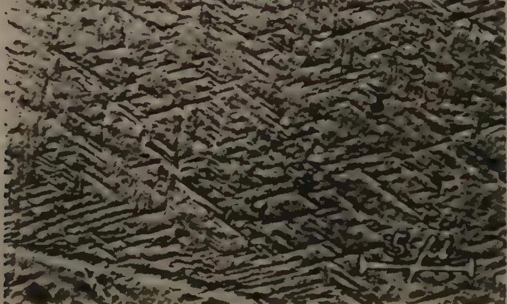



Fig. 10 Probable form of phase relationship in system ilmenite-hematite:
$M$: Minakami (original) sample
$G$: Gokurakuzi (2) ($T_s>100$°C, original) sample
$A$: Asama (original) sample
$I$: Ore-ilmenite (original) sample
$M'$: Minakami (600°C~700°C, AIR) sample

a

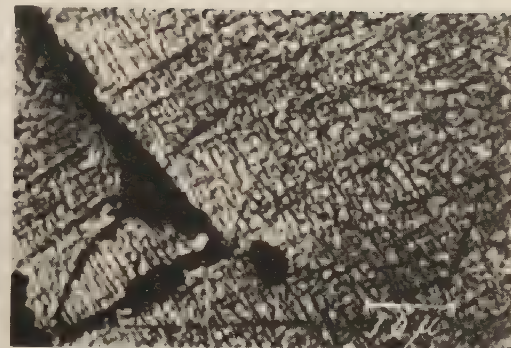
b



c

Plate 1: Electron-micrographs of polished surfaces of ferromagnetic ilmenites having the RTRM characteristics; surfaces were etched with HF for 1 minute.
a) *Sokota* (700°C AIR, 10 min.)
b) *Himesima* (660°C, AIR, 10 min.)
c) *Gokurakuzi* (1) ($T_s>100$°C, 650°C, AIR, 10 min.)


a



b

c

Plate 2: Photo-micrographs of polished surfaces of ferromagnetic ilmenites, etched with HF for 1 minute.
a) Haruna (original) sample
b) Asama (original) sample
c) Ore-ilmenite (original) sample
The well developed lamellae can be seen in all the *ferromagnetic ilmenites.*

developed lamellae were observed in all the ferromagnetic ilmenites but scarcely in the ore-ilmenite. It has, so far, been impossible to confirm the hypothesis of the "partial exsolution" by observing the differences in the occurrence of the lamellae before and after the heat treatment, because of the great diversity from grain to grain in each sample which prevented us from quantitative observation.

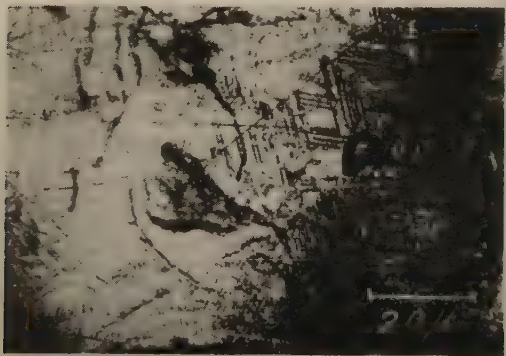
The hypothetical mechanism discussed above seems to be capable of explaining, at least qualitatively, various experimental results obtained so far. But since this mechanism presupposes many unproved conditions, it should be emphasized that it is still quite tentative. Detailed studies which may furnish more quantitative and convincing facts are required.

Whether the above mechanism is right or not, the fact that TRM characteristics of the ferromagnetic ilmenites are extremely sensitive to the thermal treatment suggests the possible usefulness of this matter as a geological thermometer that can tell not only the crystallization temperature of the minerals concerned but also their cooling rate with considerable accuracy.

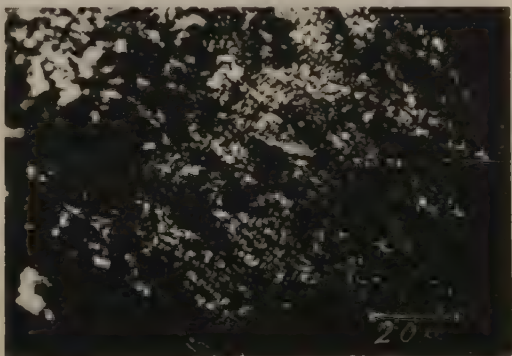
## 6. Increase of the Normal TRM by Heat Treatment at 750°C–850°C in the Open-air Tube.

In this section, the second important effect of the heat treatment on TRM,

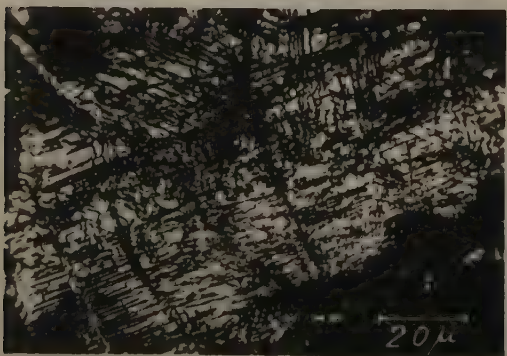
namely the intensification of the normal TRM due to the treatment at 750 C–850 C in the open-air tube, found in the samples of the ***intermediate*** $T_c$ ***and low*** $T_c$ ***groups*** (Fig. 3, c, d, $d_1$, $d_2$, e, f, g.), will be examined. The results of chemical analysis of the Haruna, Minakami and Ore-ilmenite samples before and after the treatments wihch are plotted in Fig. 4, a, d, e show the extent of oxidation occurring during the treatments. In these figures, the points representing the chemical compositions of the treated samples are well located on the theoretical oxidation lines. The X-ray analyses by Norelco were also done and the weak and broad peaks of titan-hematite and rutile were detected in addition to the intense peaks of ferromagnetic ilmenite (also broadened): the chemical compositions of these coexisting phases, estimated from the X-ray data, are also plotted in Fig. 4, a, d, e. Thus, it may be concluded that the natural ferromagnetic ilmenite and the ore-ilmenite undergo an appreciable degree of oxidation through the heat treatment at 750°C–850 C in the open-air tube for 10 minutes,* and a certain amount of titan-hematite and rutile are formed while the remaining ferromagnetic ilmenite itself becomes richer in hematite content. We named this oxidation "the oxidation of the first degree" as compared with "the oxidation of the second degree" explained in **7**. Plates 3, a, b, c are the photo-micrographs of the polished surfaces of the ***Haruna*** (850°C, AIR, 1 hr.), ***Minakami*** (820°C, AIR, 10 min.) and *Ore-ilmenite* (850°C, AIR, 10 min.) samples. Somewhat irregular pat-



a



b




c

Plate 3: Photo-micrographs of polished surfaces of samples heat treated in an open-air tube at 850°C.
a) *Haruna* (850°C, AIR, 1 hr.) etched with HF for 1 minute.
b) *Minakami* (820°C, AIR, 10 min.) etched with HF for 5 sec.
c) *Ore-ilmenite* (850°C, AIR, 10 min.) etched with HF for 5 sec.
Somewhat irregular black lamellae are titan-hematite produced by oxidation of the first degree.

* The duration of heat treatment was 1 hr. for the Haruna sample as stated in **5**.

terns of the titan-hematite produced by the oxidation of the first degree and the remaining ferromagnetic ilmenite are seen.

Taking these facts into consideration, it is possible to interpret the behaviour of TRM characteristics concerned in the following manner: Titan-hematite is known to be very weakly ferromagnetic [20]~[22]. Therefore, the titan-hematite and the rutile developed during the oxidation of the first degree will act as the non-magnetic inclusion in the ferromagnetic medium of ferromagnetic ilmenite that hinders the free movement of the magnetic domain wall, so that the coercive force of the sample will be increased. If the coercive force is increased, the intensity of the TRM is also expected to increase. However, in the case of the samples of the *high $T_c$ group*, such as the Haruna sample, the original chemical composition of them being near the limit of the ferromagnetic region indicated by the dotted line in Fig. 6, the remaining "ferromagnetic ilmenite" part itself becomes a titan-hematite by the oxidation of the first degree as shown in Fig. 4, a. This is the reason why the TRM of the *high $T_c$ group* samples does not increase in the normal direction.

To verify the above described reasonings on the mechanism of increase of TRM by the heat treatment in the open-air tube at 750°C–850°C, an additional experiment on the Minakami (40°C$>T_s>$25°C) sample was made: we measured, besides the intensities of TRM, the magnetic susceptibility $\chi$, the thermal change of saturation magntization $I_s(T)$, and the coercive force at room temperature $H_c(T_0)$ for the original

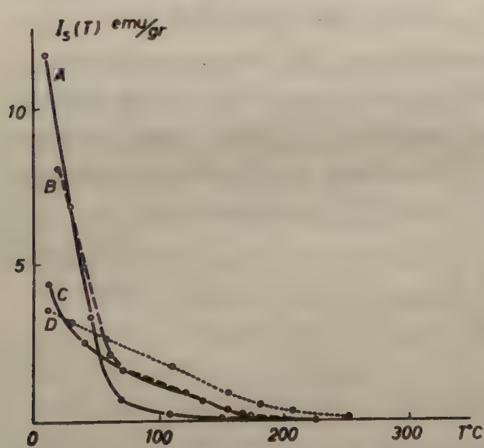



Fig. 11, a *Is* (*T*) curves of Minakami sample:
A): Minakami (40°C$>T_s>$25°C, original) sample
B): *Minakami* (40°C$>T_s>$25°C, 760°C, AIR, 10 min.)
C): *Minakami* (40°C$>T_s>$25°C, 820°C, AIR, 10 min.)
D): *Minakami* (40°C$>T_s>$25°C, 1200°C, VAC, 10 min.)
In the cases B) and C), $I_s(T_o)$ decreases by decrease in the ferromagnetic fraction, in spite of increase in Curie point. Curve D) is attributable to $I_s$ ($T_o$) of the Ti-Mt produced by reduction.

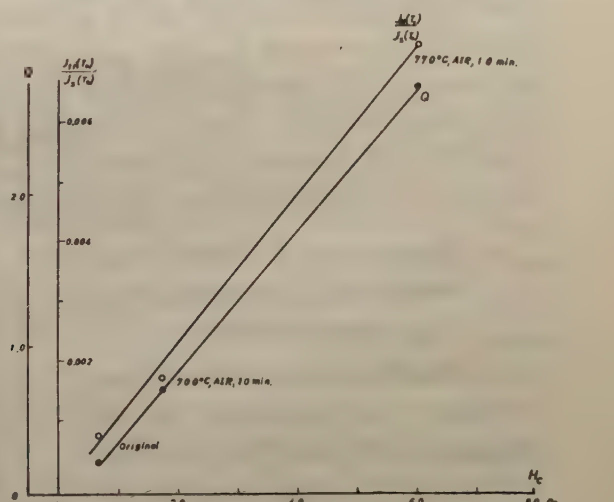



Fig. 11, b Linear relation between TRM characteristics and $H_c$ ($T_o$):
hollow circles: the ratio$=Jtr$ ($T_o$)/ $J_s$ ($T_o$),
full circles: the Q-ratio$=Jtr$ ($T_o$)/$\chi$($T_o$) $H$,
sample: Minakami ferromagnetic ilmenite (40°C$>T_s>$25°C), $H$ applied$=$2.0 Oe.

and heat treated samples. The summarized results of these measurements are shown in Fig. 11, a, b. In Fig. 11, a, the rise of Curie point is due to the oxidation of the remaining ferromagnetic ilmenite, and the decrease in the saturation magnetization at $T_0$ is due to the fact that the decrease in the fraction of the ferromagnetic substance caused by the production of titan-hematite and rutile overcomes, at $T_0$, the increase of the satuation magnetization accompanying the rise of Curie point.* Fig. 11, b shows the *Q-ratio*$=J_{tr}(T_0)/\kappa(T_0)H$, where $H=0.5\,Oe$. and the ratio: $J_{tr}(T_0)/J_s(T_0)$ as dependent on the coercive force $H_c(T_0)$, where $\kappa$, $J_{tr}$ and $J_s$ denote the susceptibility, TRM and saturation magnetization respectively. From this figure, the following relation will be established:

$$J_{tr}(T_0)/J_s(T_0)\propto H_c(T_0). \tag{1}$$

In a recent paper [11], Néel, considering the mechanism of the production of TRM, deduced the relation,

$$J_{tr}(T_0)/J_s(T_0)\propto \sqrt{H_c(T_0)}. \tag{2}$$

Although the relation (1) seems to contradict the relation (2), this by no means invalidates the manner of Néel's reasoning followed in § 57 of his paper. In the case considered by Néel, $J_s$ is assumed to vary as $(T-T_c)^{1/2}$, which is nearly true for the common ferromagnetics. But in the present case, $J_s$ varies more likely as $(T-T_c)$. This linear mode is known to be a characteristic of the ferromagnetic ilmenites. Therefore, if we accept the linear mode of thermal change of $H_c$ as reported by Nagata and Akimoto [22], we can easily show that Néel's consideration leads to the relation (1). From the point of view of clarifying the mechanism of TRM, it will be worth trying to conduct similar experiments on the Ti-Mt series, for which the parabolic mode of $J_s(T)$ is established, to examine if the relation (2) holds instead of (1).

### 7. Disappearance of TRM by Heat Treatment at $T>850°$C in the Open-air Tube.

The diminishing or disappearance of TRM of all samples by the heat treatment at $T>850$°C in the open-air tube seen in Fig. 3 can simply be attri-

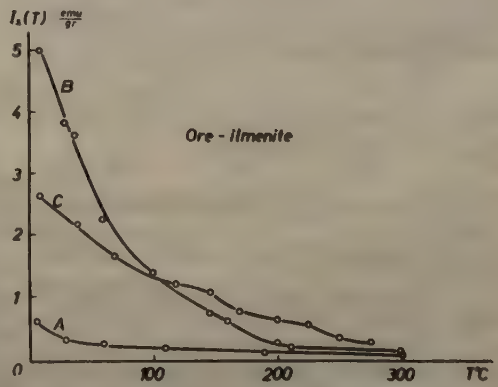



Fig. 12 $I_s$ $(T)$ curves of the ore-ilmenite samle:
A): Ore-ilmenite (original) sample,
B): *Ore-ilmenite* (850°C, AIR, 10 min.),
C): *Ore-ilmenite* (1200°C, AIR, 10 min.).
Increase in $I_s$ $(T_0)$ in B) is remarkable.

* In the case of the Ore-ilmenite, the saturation magnetization at $T_0$ was found to increase by the heat treatment as shown in Fig. 12. This means that the increase in $I_s$ of the ferromagnetic part overcame, in this case, the decrease in its fraction. Therefore, the tremendous increase in the TRM of the Ore-ilmenite in Fig. 3, g should partly be attributed to the increase of $I_s$ itself.

buted to those of the ferromagnetic nature of the samples. The diminishing or disappearance of the ferromagnetic properties has been proved to be due to the radical oxidation as shown, for example, by the chemical analyses on the Haruna, Himesima and the Ore-ilmenite samples after heat treated at 1200 C in the open-air tube (see Fig. 4, a, b, e). The results of the X-ray analyses showed that these treated samples consist of pseudobrookite ($Fe_2TiO_5$), titan-hematite and rutile as indicated by the dotted lines in the figures. We named this oxidation, which produces the pseudobrookite phase, "the oxidation of the second degree." Recently, we have been informed that Curnow and Parry also have found two stages of oxidation of ilmenite which are completely equivalent to our own results [25]. Plate 4 is a photo-micrograph of the polished surface of the *Ore-ilmenite* (1200°C, AIR, 10 min.) sample. The coarse coexistance of the pseudobrookite phase and the titan-hematite phase can be observed.

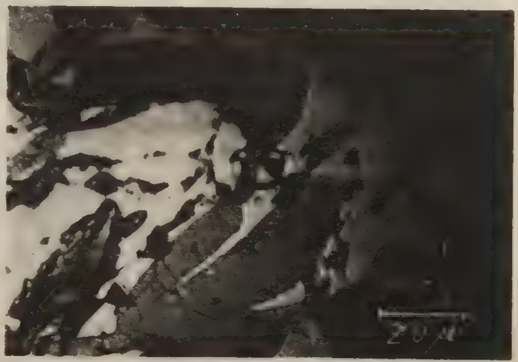
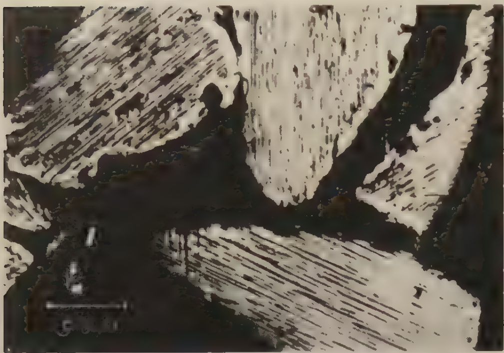


Plate 4: Photo-micrograph of polished surface (not etched) of the *Ore-ilmenite* (1200°C, AIR, 10 min.) sample. Darker part is pseudobrookite produced by oxidation of the second degree, and the lighter part is the remaining Il-Ht series.

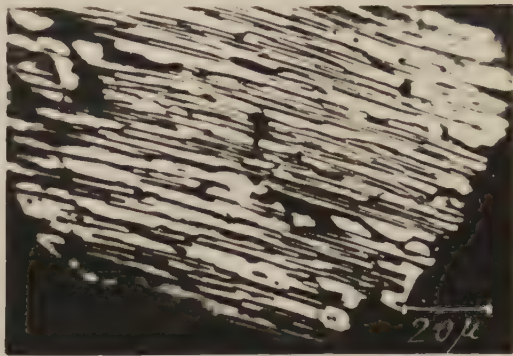
### 8. Intensification of Normal TRM by the Heat Treatment at $T \gtrsim 800$ C in the Evacuated Sealed Tube.

As already mentioned, the effect of heat treatment at 600°C–700°C can be interpreted by the hypothesis of the "partial exsolution" (5). Therefore, we examine, in this section, the causes of the intensification of the normal TRM due to the heat treatments at $T \gtrsim 800$°C in the evacuated sealed tube. The intensification was found for all the samples except the Asama sample (see Fig. 3, e). The chemical compositions of some of the samples heat treated at 1100 C and 1200°C in the evacuated sealed tube are shown in Fig. 4 by which it is verified that considerable reduction actually took place. Through X-ray analysis, it was also ascertained that the treated samples now contain some members of the Ti-Mt series. The chemical compositions of both the Ti-Mt and the remaining Il-Ht estimated from the X-ray data are also plotted in Fig. 4. It is clearly seen from these figures that the more the reduction proceeds, the more the chemical compositions of both Ti-Mt and Il-Ht get close to those of pure ulvöspinel and ilmenite. Even when Ti-Mt was not identified by X-ray analysis because of its scanty content as in the case of the *Ore-ilmenite* (1200°C, VAC, 10 min.), microscopic observations revealed its existence in some grains as can be seen in Plates 5, a, b, c. In these photo-micrographs, Ti-Mt can be observed as thin parallel layers of uniform direction. This unidirectionality of Ti-Mt in the host of the Il-Ht series is in sharp contrast to the tridirectional occurrence of ilmenite in magnetite.

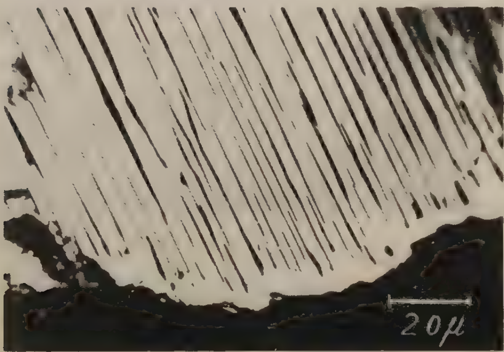
Now, taking the Curie points of the Ti-Mt and the Il-Ht shown in Table II and Fig. 4 into account, we may be able to conclude that the intensification in the normal



a




b




c

Plate 5 : Photo-micrographs of polished surfaces of samples heat treated in evacuated sealed tube at 1200°C. Surfaces were etched with HF for 5 sec.

a) *Haruna* (1200°C, VAC, 1 hr.)

b) *Minakami* (1200°C, VAC. 10 min.)

c) *Ore-ilmenite* (1200°C, VAC, 10 min.)

The black lamellae are Ti-Mt produced by reduction.

TRM dealt with in this section is attributed to the appearance of Ti-Mt with the Curie point higher than $T_0$ in the sample. It will be noteworthy, moreover, that at the same time the remaining Il-Ht series becomes non-magnetic at $T \gtrsim T_0$. Therefore, in these cases, the TRM is generated entirely by the newly developed Ti-Mt, while the Il-Ht remainders which are now non-magnetic act as an inclusion as did rutile and titan-hematite in the cases treated in **6**. Figs. 11, a and 13 show the $I_s(T)$ curves of some of the treated samples, such as the *Minakami* (1200°C, VAC 10 min.), *Haruna* (1100°C, VAC) and *Sokota* (1100°C, VAC) samples. These curves are quite reasonably interpreted as those of the Ti-Mt. In the case of the Asama sample, for which no increase of TRM was observed (Fig. 3, e), it may be considered that both of Ti-Mt and Il-Ht are made non-magnetic because of radical reduction that takes place (cf. **10**).

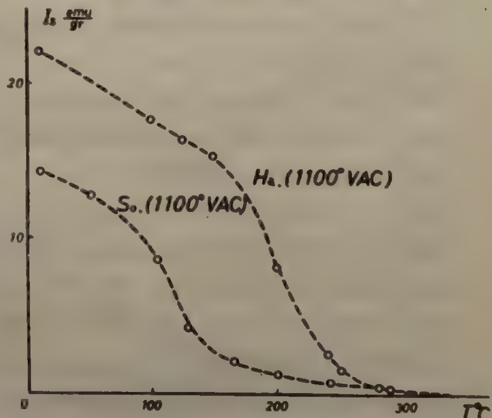



Fig. 13 $I_s$ ($T_o$) curves of Haruna and Sokota samples, heat treated in evacuated sealed tube at 1100°C for 10 minutes : the curves are attributable to those of the Ti-Mt produced by reduction.

## 9. Production of RTRM by Heat Treatment at 1100°C in the Evacuated Sealed Tube, Found for the Ferromagnetic Ilmenite of Gokurakuzi (1) ($T_s$ > 100°C).

The reversal of TRM of the *Gokurakuzi* (1) ($T_s$ > 100°C, 1100°C~1200°C, VAC, 10 min.) seen in Fig. 3, $d_1$ is the only case where the heat treatment in the evacuated sealed tube produced RTRM. As was seen in **8**, the heat treatment at 1100°C~1200°C in the evacuated sealed tube causes a certain amount of reduction (see Fig. 4, c), through which Ti-Mt is produced. Fig. 14 shows the $I_s(T)$ curve of this treated sample. In this curve, it will be noticed that both the Ti-Mt and the remaining Il-Ht are ferromagnetic: their Curie points being about 450°C and 90°C respectively. This result is in good agreement with the X-ray data.

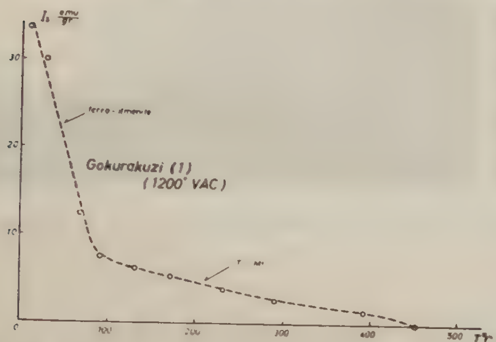



Fig. 14 $I_s(T_o)$ curve of *Gokurakuzi* (1) ($T_s$ > 100°C, 1200°C, VAC, 10 min.) sample, showing RTRM: the coexisting ferromagnetic ilmenite and Ti-Mt, brought about by reduction, are considered to interact magnetically to produce the RTRM.

Judging from these data, we may be able to interpret this type of RTRM as caused by the magnetic interaction between the newly formed Ti-Mt and the remaining ferromagnetic ilmenite. Haruna-type RTRM was at first assumed to be caused by the magnetic interaction of this kind [4]~[7], [15], but this was shown to be false and interaction was attributed to that between two ferromagnetic ilmenites as has already mentioned [12]. Hence, the RTRM of the *Gokurakuzi* (1) ($T_s$ > 100°C, 1100°C ~ 1200°C, VAC, 10 min.) should be regarded as the first example of RTRM originating from the magnetic interaction between Ti-Mt and ferromagnetic ilmenite. The state of the coexistence of these two phases observed in each grain is shown in Plate 6.

Plate 6: Photo-micrograph of polished surface of *Gokurakuzi* (1) ($T_s$>100°C, 1200°C, VAC, 10 min.) sample, etched with HF for 5 sec. The black lamellae are Ti-Mt produced by reduction. This sample shows RTRM.

## 10. Disappearance of TRM by Heat Treatment in the Evacuated Sealed Tube.

When the reduction during the heat treatments in the evacuated sealed tube proceeds far enough, both of the newly formed Ti-Mt and the remaining Il-Ht series become non-ferromagnetic at room temperature. In such a case, the Ti-Mt becomes ulvöspinel and the Il-Ht becomes ilmenite. Examples of such cases were furnished by the Himesima and Asama samples: (Fig. 4. b, e and Table II). Thus, we may be able to attribute the observed disappearance of TRM of these


a b

Plate 7 : Photo-micrographs of polished surfaces of samples which underwent radical reduction by heat treatment in evacuated sealed tube. Surfaces were not etched.

a) *Himesima* (1100°C, VAC, 10 min.)

b) *Asama* (1100°C, VAC, 10 min.)

Reduction taken place was so radical that the main part (darker) is ulvöspinel, in which ilmenite remains in lamellae (lighter).

samples to the disappearance of ferromagnetism due to extreme reduction. Plates 7, a, b are the photo-micrographs of these samples, where the darker part is ulvöspinel and the lighter lamellae are ilmenite.

### 11. Magnetic Interaction between Titano-magnetites (Ti-Mt).

In the preceding sections, we have dealt with the role of magnetic interaction between different minerals in the course of the development of TRM, including the case where one of the two minerals was non-ferromagnetic. The samples dealt with so far have been restricted to those derived from the natural ferromagnetic ilmenites by various heat treatments.

In this section, we will discuss the experimental results concerning the effect of the magnetic interaction between two Ti-Mts. The first example is the result of the study on the anomalous increase of TRM during its thermal demagnetization. This phenomenon has been found on the Ti-Mts of the Haruna pumice (of which the ferromagnetic ilmenite produces the Haruna-type RTRM) and the iron sand from the biotite rhyolite pumice of Niisima [3] [15]. In the following we will quote the results of the detailed examination carried out by Nagata and Ozima [16]. When the TRM of certain Ti-Mt samples as mentioned above is thermally demagnetized in a non-magnetic space, it shows an anomalous increase at a temperature a little below the Curie point, as schematically illustrated in Fig. 15 (curve C). This phenomenon has been called " the anomalous increase " because the ordinary TRM is known to decrease monotonously during the process of thermal demagnetization [15]. The origin of this phenomenon was also assumed to be a magnetic interaction between two ferromagnetic constituents in the sample : the outline mechanism will be easily seen from the schematic representation in Fig. 15 [15], [16]. To prove this mechanism Nagata and Ozima made the following study.

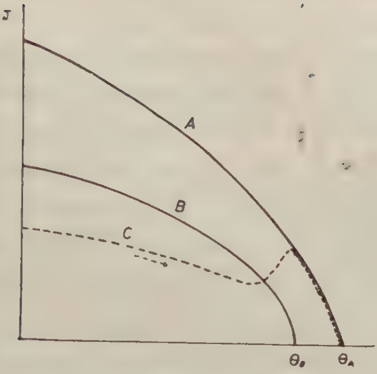



Fig. 15 Schematic representation of the "AI" (anomalous increase) phenomenon of TRM: curves $A$ and $B$ are TRMs of the $H$- and $L$-constituents: if the TRM of the $L$-constituent is produced in the reverse direction by magnetic interaction with the TRM of the $H$-constituent, the resultant TRM of the composite system will become as curve $C$. (after Nagata and Ozima).

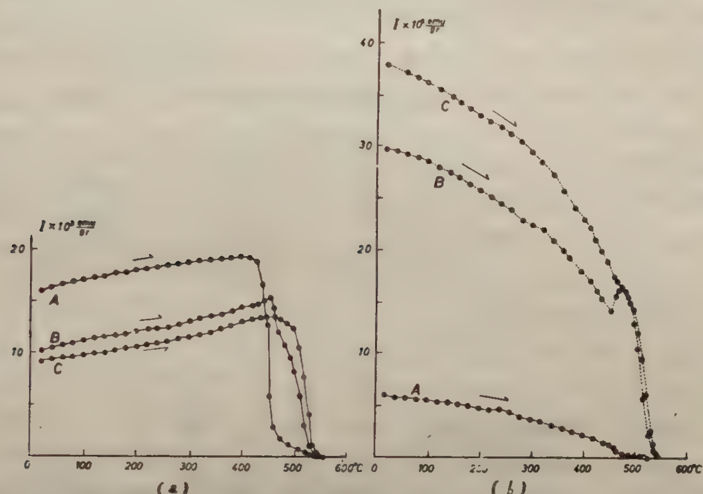



Fig. 16, a Thermal change of reversible magnetization $x(T)H$ of Niisima Ti-Mt sample: $H$=2.0 Oe.

b Thermal decay mode of total TRM of Niisima Ti-Mt sample: $H$=2.0 Oe.

A): Niisima (original)

B): *Niisima* (650°C, AIR, 3.5 hrs)

C): *Niisima* (650°C, AIR, 12 hrs.)

In B), coexistence of $H$- and $L$-constituents with Curie point at 470°C and 540°C can be observed in Fig. 16, a, while "AI" phenomenon is observed in Fig. 16, b. (after Nagata and Ozima)

It was discovered that this phenomenon is sensitively dependent on the heat treatment of the sample as in the cases treated in the previous sections. For example, Fig. 16, a, b show that the Niisima sample, originally having no "anomalous increase," reveals a remarkable "anomalous increase" after the heat treatment at 650°C in air for 3.5 hours. From the curves in Fig. 16, a, it may be concluded that the Curie points of the two constituents are 540°C and 470°C respectively, and that the constituent having the higher Curie point ($H$-constituent) is developed in the host with the lower Curie point ($L$-constituent) by the heat treatment. It was also ascertained that the development of the $H$-constituent is caused by the oxidation of the $L$-constituent in a form of fine lamellae. Fig. 16 also shows that when oxidation

proceeds far enough, all of the $L$-constituent is transformed into the $H$-constituent so that the "anomalous increase phenomenon" disappears again. If the above quoted two-constituent model is true, it can be said that there should be the possibility of producing a total reversal of TRM, which is equal to the very strong "anomalous increase phenomenon," by some suitable heat treatment. This also means that a reversal of natural remanent magnetism of Ti-Mts may have possibly been produced in nature provided some geological conditions favoured the appropriate oxidation.

Another important result on the magnetic interaction between Ti-Mts has been reported in a series of works by Kawai, Kume, Sasajima and Yasukawa [17]~[19]. Their work can be summarized briefly as follows: They postulate that the coexistence of various Ti-Mts with different Curie points in rocks has been brought about by exsolution that has been taking place since the time of the crystallization of the rocks. On the other hand, they have reported that, in some rocks, the natural remanent magnetism changes its sign, sometimes even twice, in the course of thermal demagnetization at the temperatures corresponding to the Curie points of Ti-Mts contained. They interpret this phenomenon in terms of magnetic interaction: let the original phase of a Ti-Mt in a rock be represented by $P$ in Fig. 17, the phase diagram proposed by these authors [17], [27]. This Ti-Mt undergoes an exsolution as the temperature falls. The degree of exsolution is determined by the cooling rate and the age of the rock. Therefore, on the average, Ti-Mts in older rocks should be more completely dissolved into two phases. They illustrated this matter by examining some 400 natural Ti-Mt samples. Now, one can define a temperature at which the exsolution took place most effectively by the precise measurement of thermo-magnetic curve, provided the solvus relation in Fig. 17 is correct. If the phases found by the thermo-magnetic study are $H_1$ and $L_1$, the effective aging temperature is $T_1$. Since the Curie point of this system can be expressed by the straight line $MN$ in Fig. 17, the Curie points of $H_1$ and $L_1$ are lower than $T_1$, then natural remanence produced during further cooling will be a stable TRM. In such a case, however, the TRM of $L_1$ and the remaining $P$ may be reversed if the magnetic interaction with $H_1$ is strong enough. If the effective aging temperature is $T_2$ in Fig. 17, the natural remanence of $H_2$ would not be a stable TRM because the Curie point of $H_2$ is higher than $T_2$, and may be directed reversely if the magnetic interaction with the already magnetized $P$ is strong enough. The natural remanence of $L_2$ may also be reversed for the same reason. Further, if the effective aging temperature is as low as $T_3$, the remanent magnetism of both $L_3$ and $H_3$ would not be a

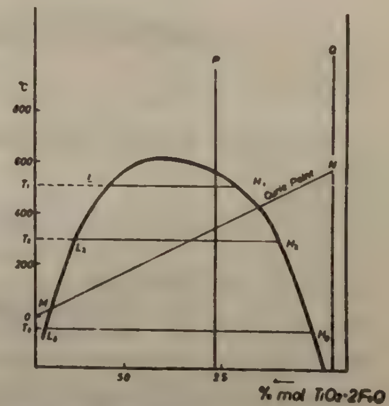



Fig. 17 Probable form of phase relationship in system magnetite-ulvöspinel, illustrating the exsolution process through which natural titanomagnetites are formed. (after Kawai)

stable TRM and may be reversed. Therefore, in general, the remanent magnetism of the older rocks, in which the equilibrium state at the ambient temperature is fairly completely established, may not be the stable TRM and hence could not be used for palaeomagnetic purposes.

On this point, the present author would like to propose that the palaeomagnetic study may be still permissible if the Ti-Mts concerned are as small in Ti content as the one expressed by $Q$ in Fig. 17, because such pure magnetites should have been free from any secondary phase transformation however old they may be. Occurrence of such a pure magnetite in igneous rocks, however, may be rather rare. These considerations indicate that the natural remanent magnetism of such a pure magnetite as found in some metamorphic rocks or ores will be of much importance in future studies in the field of palaeomagnetism.

## Conclusion

The role of magnetic interaction between ferromagnetic minerals in the course of the production of TRM has been examined experimentally on the system $FeO$-$Fe_2O_3$-$TiO_2$. Since the magnetic interaction is expected to be dependent on the various magnetic properties and the spatial relationships of the minerals concerned, we attempted to examine as many combinations of minerals as possible by means of heat treatments on the natural ferromagnetic minerals. Our present work, however, was confined to the minerals derived from the natural ferromagnetic ilmenites, so that we have referred to the works of other investigators of the titanomagnetites. The general conclusions obtained may be summarized as follows:

1) When two ferromagnetic minerals coexist within a grain in the form of alternately laminated structures, magnetic interaction is negative. The most typical of this case is the Haruna-type RTRM, in which the two participating minerals are both ferromagnetic ilmenites. The occurrence of the two ferromagnetic ilmenites with different but similar properties in rocks was interpreted as caused by the "partial exsolution," brought about by the rapid cooling of the mother rock.

The RTRM of this type is very sensitive to the heat treatment of relatively short duration. The most typical of this is the artificial production of the Haruna-type RTRM by heat treatment on some ferromagnetic ilmenites which originally show only a normally directed TRM.

An example of RTRM caused by the magnetic interaction between titanomagnetite and ferromagnetic ilmenite was also found in the present work. This RTRM was produced on a natural ferromagnetic ilmenite by heat treatment in an evacuated sealed tube, and was interpreted as caused by the interaction between the titanomagnetite developed by the reduction process and the originally existed ferromagnetic ilmenite.

The "anomalous increase" of TRM and the reversal of natural remanence during the process of thermal demagnetization revealed in some titanomagnetites were referred to as examples of the phenomena caused by similar magnetic interactions.

2) Another remarkable fact is that when non-ferromagnetic minerals, such as

titan-hematite, rutile, ilmenite or ulvöspinel, are intergrown with the host of ferromagnetic one, both the coercive force and the TRM of the sample are intensified. The increase of the coercive force is attributed to the development of the finely intergrown non-ferromagnetic materials which hinder the free displacement of the magnetic domain walls in the sample. The increase of the intensity of TRM may be related to the corresponding increase in the coercive force as Néel suggested. An empirical relation,

$$J_{tr}(T_0)/J_s(T_0) \propto H_c(T_0),$$

has been obtained for the heat treated ferromagnetic ilmenite containing titan-hematite and rutile.

Although the principal purpose of the present study was to examine the role of magnetic interaction in the rock's magnetism, the experiments conducted in this work revealed some interesting results on the oxidation and the reduction of ilmenites, both ferromagnetic and non-ferromagnetic: 1) the oxidation of ilmenites by heating in air proceeds in two steps. The oxidation of the first degree takes place effectively below 850°C, and produces some titan-hematite and rutile in the sample. The Curie point of the remaining ferromagnetic ilmenite is raised and the originally non-ferromagnetic ilmenite is made ferromagnetic. The oxidation of the second degree takes place when the temperature exceeds 850°C and produces some pseudobrookite in addition to titan-hematite and rutile. The remaining ferromagnetic ilmenite itself is also oxidized to become non-ferromagnetic. 2) the heat treatment in a sealed quartz tube evacuated to as low as $10^{-3}$mmHg brings about some reduction, and the ferromagnetic ilmenites become poorer in hematite content while some titanomagnetite is produced. When reduction proceeds radically, the final products are nearly pure ulvöspinel and ilmenite, both being non-ferromagnetic.

**Acknowledgment**

The writer would like to thank Prof. T. Nagata for his kind suggestions and the most pertinent directions throughout the course of this study. He also cordially acknowledges the wholehearted encouragement rendered by Dr. T. Rikitake. The idea of the partial exsolution of ferromagnetic ilmenites was first suggested by Prof. H. Kuno, and all the chemical analyses of the samples were conducted by Mr. K. Katsura of the Tokyo Institute of Technology, to both of whom the writer is extremely indebted. The writer's cordial thanks are due to Mr. K. Akashi who kindly allowed the writer to use the Akashi Electron Microscope in his factory, and to the members of the Akashi Seisakusho, especially Miss. E. Iguchi, for their assistance in microscopy. He also wishes to thank Messrs. N. Kawai, S. Kume, S. Sasajima and K. Yasukawa for referring to their works. His most sincere thanks are due to his senior colleague Mr. S. Akimoto whose work on the fundamental properties of ferromagnetic minerals is the basis for the present study. Finally, the author cordially acknowledges Messrs. S. Aramaki and S. Tono of Geological Institute, Tokyo University for their valuable opinions and for the assistance rendered in the microscopic study: Messrs. Y. Shimizu,

M. Ozima, K. Kobayashi and H. Maekawa of the Geophysical Institute, Tokyo University, for their useful discussions and for their assistance in experiments and in the preparation of the figures for the present paper: Miss. S. Sato for her kind help in the computation of X-ray data.

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# Observation of Cosmic-Ray Neutron Intensity at Geomagnetic Latitude 25°N

## Part II. 27-Day Recurrence Tendency and Solar Activity

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(Read May 14, 1956; Received Aug. 14, 1956)

**Abstract**

A remarkable 27-day recurrence tendency of cosmic-ray neutron intensities was noticed through the observation at Nagoya (geomagnetic latitude 25°N, sea level) from October, 1954 to July, 1955. This tendency is completely in the same phase with that of the solar activity during the same period. This phenomenon seems to show that some part of low energy cosmic rays are usually emitted from the sun.

### 1. Introduction

Many people [1] have hitherto investigated the 27-day recurrence tendency of cosmic rays from the various points of view. The results thus obtained could be generally summarized as follows.

1. There is close correlation between the recurrence of cosmic rays and that of magnetic charactor figure.

2. In general, the phases of the recurrence of these two quantities are not the same with each other, and in some cases the phase difference reaches to a half period.

3. There is similar relation as that mentioned above between cosmic rays and solar activity, or sunspot relative numbers.

The interpretation [2] recently proposed for these facts is the view that a magnetic cloud, is emitted from a sunspot region, and it makes the cosmic-ray intensity decrease and gives some effects on geomagnetism simultaneously. If so, the cosmic-ray intensity should decrease corresponding to the appearance of active solar phenomena through their 27-day recurence.

The following facts suggest that another phenomenon must be taken into account in addition to the phenomenon mentioned above.

### 2. Results

The data used for this analysis are shown in Table I.

The observation of neutron intensity and its atmospheric effect were described in Part I [3]. The period used for the present analysis is from October, 1954 to July,

* Now at Scientific Research Institute, Tokyo.

*Table I. List of the data used*

| Data | Station | Geomagnetic latitude | | Notation |
|---|---|---|---|---|
| Neutron intensity | Nagoya | 25°N | Corrected for pressure | $I_n$ |
| Meson intensity | Tokyo | 25.5°N | Corr. for pressure and temperature | $I_m$ |
| Solar radio noise | Toyokawa | 25°N | Observed frequency 3750Mc/s | *S.N.* |
| Sunspot relative numbers | Tokyo | 25.5°N | | $R$ |

1955. The daily means of neutron intensities ($I_n$) and solar noise (*S.N.*) in this period are plotted in Fig. 1, respectively. It will be easily seen from this figure that there are both 27-day periodic variation and longer term variation than it.

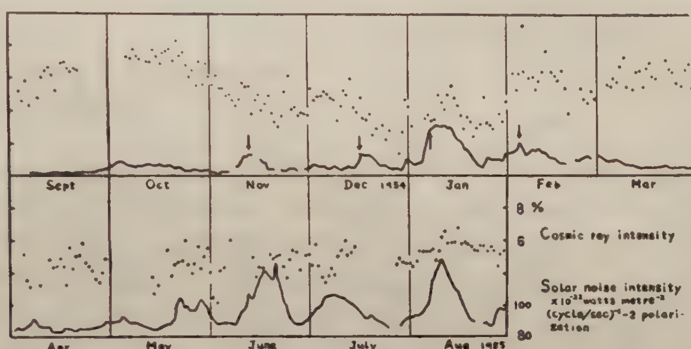


Fig. 1. Daily variations of neutron intensity and solar noise intensity. Arrows indicate days of remarkable solar noise intensity.

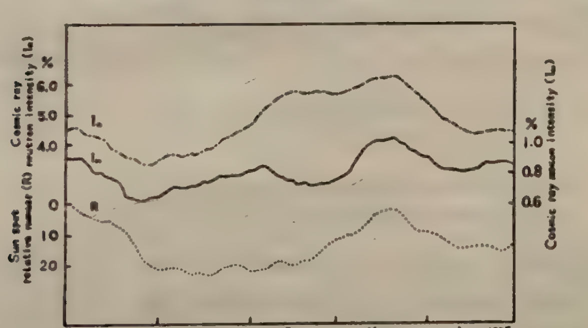


Fig. 2. Daily variations of 27-day running average of cosmic-ray intensities and solar activity.

In order to pick up the latter variation only, 27-day running average was drawn in Fig. 2, in which sunspot relative numbers ($R$) was used in stead of solar noise. Cosmic-ray intensities, both neutron and meson components, have negative correlation with $R$. The correlation coefficients obtained for every month are shown in Table II.

To pick up the 27-day periodic variations, the intensity deviations from 27-day running averages were calculated for every day. These deviations were superposed by Chree's method, where the days of remarkable solar noise shown by arrows were adopted as "epoch" days. Thus, a remarkable 27-day recurrence tendency was

*Table II. Correlation coefficients among cosmic-ray intensity and sunspot relative numbers represented as 27-day running averaged values.*

| | $\gamma_{NR}$ | $\gamma_{MR}$ | $\gamma_{NM}$ | $\beta_{NM}$ |
|---|---|---|---|---|
| Nov. | —— | −0.87 | —— | —— |
| Dec. | −0.92 | −0.93 | +0.99 | +4.5±0.1 |
| Jan. | −0.45 | −0.10 | +0.90 | +6.7±0.5 |
| Feb. | −0.55 | +0.50 | −0.81 | −4.9±0.4 |
| Mar. | −0.82 | −0.88 | +0.67 | +1.8±0.3 |
| Apr. | −0.67 | −0.20 | +0.38 | +3.3±1.1 |
| May | —— | −0.89 | —— | —— |

$N$: Neutron, $M$: Meson, $R$: Sunspot relative numbers.

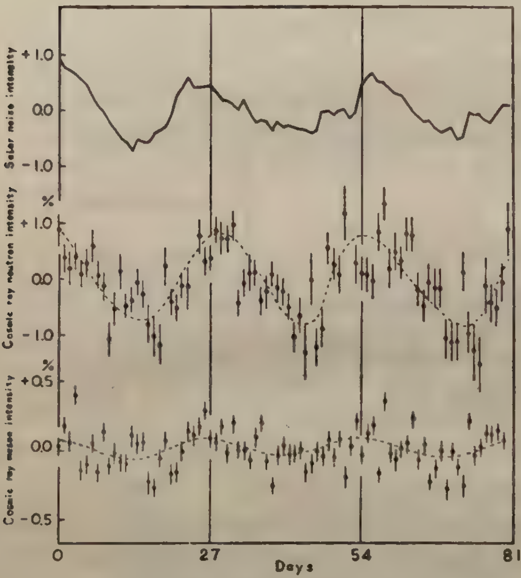


Fig. 3. 27-day recurring variations of cosmic-ray intensities and solar noise.

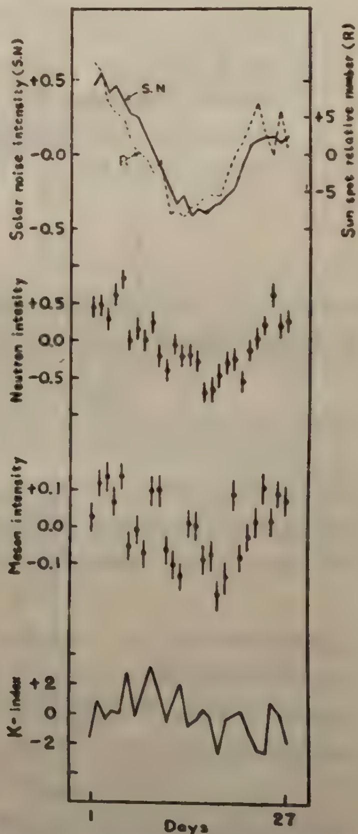


Fig. 4. 27-day recurring variations superposed on three periods in Fig. 3.

obtained as shown in Fig. 3. Table III is the results of Fourier analysis of these curves. All numerical values on the table are well significant statistically, and the advancements or delays of the phase of cosmic rays of each cycle nearly correspond to those of solar noise. As it is clear from the average curves over three periods shown together with $R$ in Fig. 4, both variations of meson and neutron intensities are perfectly in the same phase, i.e. positive correlation, with the variations of the solar activity represented by solar noise, or sunspot relative numbers $R$.

### 3. Interpretations and Discussions

We would like to adopt a most simple assumption to interprete the phenomena

*Table II. Harmonic analysis of 27-day recurrence of solar noise, neutron and meson intensities.*

| Period number | | Amplitude (%) | Phase (days) | Ratio of amplitude of $I_n$ and $I_m$ |
|---|---|---|---|---|
| I | S.N. | 7.15 | +1 | 8.8±3.1 |
| | $I_n$ | 0.53±0.06 | +2 | |
| | $I_m$ | 0.06±0.02 | +1 | |
| II | S.N. | 2.63 | +2 | 8.9±2.6 |
| | $I_n$ | 0.62±0.06 | +2 | |
| | $I_m$ | 0.07±0.02 | +3 | |
| III | S.N. | 3.27 | +4 | 6.3±2.0 |
| | $I_n$ | 0.44±0.07 | +4 | |
| | $I_m$ | 0.07±0.02 | +3 | |
| Mean | S.N. | 4.58 | +2 | 7.0±0.9 |
| | $I_n$ | 0.56±0.04 | +2 | |
| | $I_m$ | 0.08±0.01 | +2 | |

mentioned above. Regarding only to the long term variation, longer than 27 days, it is plausible to explain the relation as a modulation effect mentioned in introduction. However, we should like to suppose that cosmic-ray particles responsible for the 27-day recurrence are directly emitted from the sun, though Morrison *et al* supposed a modulation effect by the corpuscular stream from *M*-region.

The reason is as follows. About the difference between the diurnal variation during the days of maximum intensity and that of minimum intensity of cosmic rays in Fig. 4, some difference should be expected from Morrison's theory and the view of solar emission. Therefore, 7-day average diurnal variations were calculated taking the

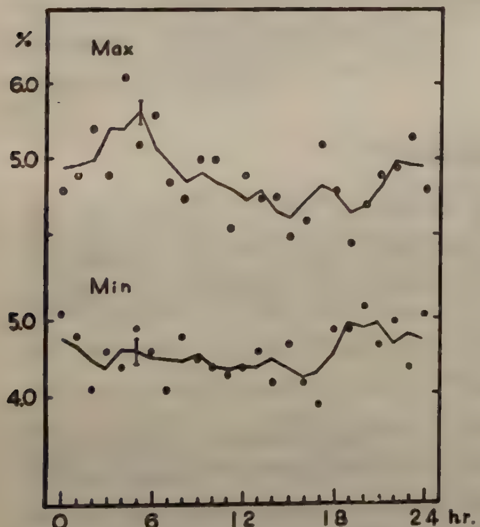


Fig. 5. Diurnal variations averaged by taking the maximum and minimum days of cosmic-ray neutron intensities in Fig. 4 as the centre. Solid curves show 3-hr running averaged values.

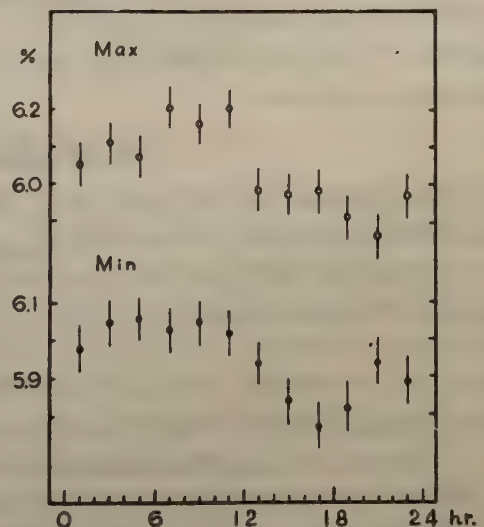


Fig. 6. Diurnal variations averaged by taking the maximum and minimum days of cosmic-ray meson intensities in Fig. 4 as the centre.

maximum and minimum days of cosmic-ray intensities as the centre, respectively. The results are shown in Figs. 5, 6. In the case of neutron, there is practically no diurnal variation for the minimum days, but a certain increase of intensity seems to exist at about 4-hr. local time for the maximum days. According to Morrison's view, primary cosmic-ray intensity should be affected uniformly over all directions by the magnetic clouds.

Therefore, it is impossible to expect that some increase appears at a special local time. If charged particles of lower energy come from a direction of the sun, it will be able to expect the increase at about 4-hr., which corresponds to a so-called impact zone. Consequently it is reasonable to consider that there was a little contribution of solar cosmic rays during this period.

As the variation of meson intensity is about a seventh of neutron variation (Table III), considerably higher accuracy will be necessary to detect above mentioned difference between diurnal variations of meson intensities.

About the 27-day periodic variation, it will be noticed that the relation obtained for the phase between cosmic ray and sunspot numbers $R$ is not the same as the facts known untill now. The period of our data just corresponds to the minimum in the eleven year change of solar activity and few cosmic ray storms happened during this period. On the other hand, most of results obtained untill now were based on the data during the periods the sunspot relative numbers was maximum or moderate. In the latter case, the effect of magnetic clouds would be so strong that the decrease of cosmic-ray intensity overtakes the increase due to solar emission.

On the contrary, it may be supposed that during this period the effect of magnetic clouds is too small to overtake the increase due to solar emission. It is too early to conclude that this phase relation will appear in every sunspot minimum. However, it can be concluded that the effect of solar emission appeared at least in this sunspot minimum.

## 4. Acknowledgements

The authors wish to express their hearty thanks to Prof. Y. Sekido for his valuable discussion and criticism throughout this work. They are also grateful to Prof. A. Kimpara and Dr. H. Tanaka at Research Institute of Atmospherics, Nagoya University for their supply of solar radio noise data.

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# Geomagnetic Variation due to the Solar Eclipse of June 20th, 1955

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(Read Oct. 29,1955, Received Sep. 5, 1956)

**Abstract**

In order to investigate the geomagnetic variaton due to the Solar Eclipse of June 20th, 1955, the $S_q$-field at $13^h30^m$ of this day is expressed by a simple, and the current-system of the additional field due to the eclipse is drawn, using the present data and a quantitative result on the bygone eclipse.

## 1. Introduction

Statistical and theoretical treatments on geomagnetic variation during solar eclipses have been attempted, but it can hardly be said that the results from both treatments show good agreement with each other. Most of statistical treatments based on observational data were unsuccessful owing to the characteristic irregularity of the $S_q$-field. The theoretical ones dealt with an ideal case and its modification [1], but the actual phenomena are too complex to be explained by an ideal expression. In order to carry out a detailed investigation of the effect of the solar eclipse of June 20th, 1955, a temporary observatory was set up at Naze (129°30'9 E, 28°22'1 N, Amami-Oshima, one of the South-western Islands in Japan), in addition to the existing observatories. The day was magnetically quiet and very favourable for detecting the eclipse-effect. The results of investigation are reported, on the basis of observed data at the several stations observing this eclipse.

## 2. Original Data and the Normal Curve

As a first step towards detecting the magnetic variation due to the solar eclipse, we must determine the normal curve (the $S_q$-variation on the eclipse-day assumed to be free from the eclipse-effect). The normal curve is conventionally determined by the mean curve of several quiet days before and after the eclipse-day. As the $S_q$-variation at the middle latitude changes its intensity and its form day by day, this mean curve does not always represent the normal curve. The author attempted to find out the eclipse effect by applying a part of normal curve for a few hours, determined from an analysis of all the available observational data around Japan.

Fig. 1 shows the vector-diagrams of the magnetic variation observed on that day at the following Japanese stations: Memambetsu (M), Kakioka (K), Aso (A) and Naze (N), and their stations are shown in Fig. 2. From Fig. 1 it can be seen that the horizontal vectors of these observatories take almost constant values (about 30 gamma and the direction is 10°～20° north from west) during two hours centred at $13^h30^m$

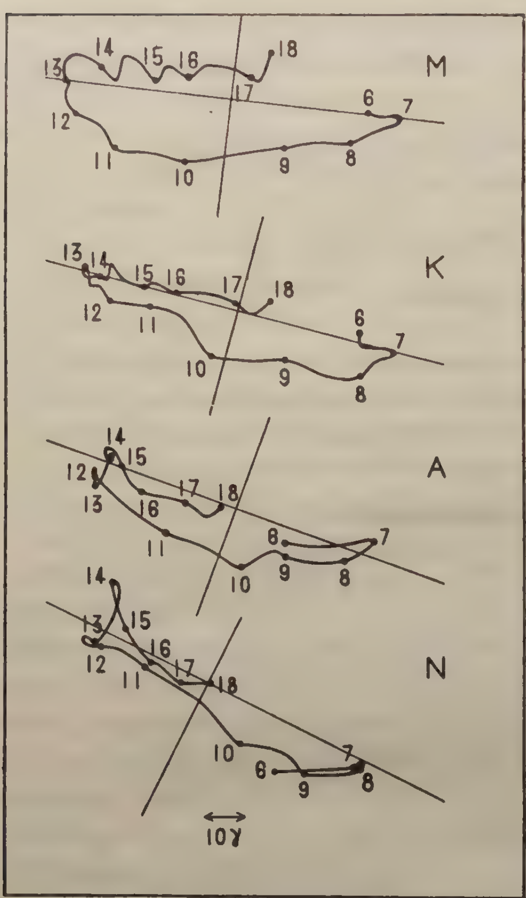



Fig. 1 Vector Diagrams

J.S.T., when the maximum obscuration occurs at this region. This suggests that the general trend of the $S_q$-field at this region is approximately uniform during these two hours. This fact is very favourable for the application of a special method for the normal curve. The general trend of the $S_q$-field at $13^h 30^m$ J.S.T. is shown in Fig. 2, using the data of the observatories in Japan and Kodaikanal in India. In this case, the normal curve of the station is easily deduced from the curves of the consecutive stations as follows: the current-system equivalent to this field is expressed by parallel current and its direction is about 10°～20° east from the Magnetic North, therefore the normal curves at these five stations can be considered as the same af five stations situated in the northern quarter of the eclipse-area. For the sake of simplicity we introduce a new coordinate-systems as parallel ($X'$) and perpendicular ($Y'$) to the $S_q$-current. The vector-diagram and the

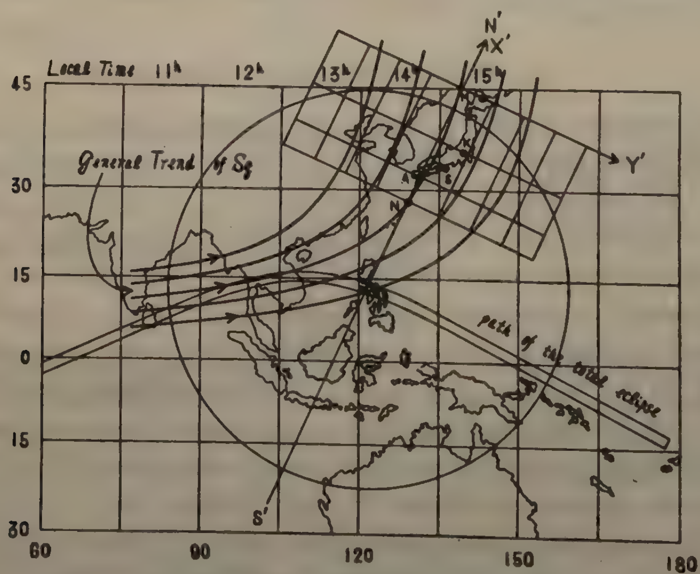



Fig. 2 General State at $13^h 30^m$ J.S.T. June 20th, 1955.

curve of Memambetsu seems to undergo so little effect of the eclipse that we may neglect it. Then the normal curves of the other stations are deduced from the Memambetsu-curve. Strictly speaking, the general trend of the $S_q$-field is not perfectly parallel and is not in a straight line for the whole region considered here. To determine the normal curve, it is necessary to introduce some corrections due to the difference between local times and the amplitude deduced from variations at the forenoon (right-side of the vector diagrams) uneffected due to the eclipse. As the $X'$-axis is parallel to the general trend to the $S_q$-field on this day, the $X'$-component of geomagnetic variation on this day could be taken as the eclipse-effect. But this is not satisfactory either, for the above mentioned reason about the general trend of the $S_q$-field. From the theoretical consideration as first shown by Chapman [1], we may assume that there are no effects at the hour of the maximum obscuration at each station. Our analyses are based on this assumption.

### 3. The Additional Field due to the Eclipse

The progressing velocity of the eclipse shadow is so slow that the additional field may be considered as a steady state. Therefore the current-system equivalent to this field is to be closed. In order to investigate the current-system of the additional field, it is necessary to have observations sufficiently distributed in the eclipse-area in a large circle as shown in Fig. 2, whose diameter is about 7,000 km. We have only five (as shown in Fig. 2 by black circles), and their distributions are arranged along the $X'$-component and not along the $Y'$. Fortunately, the $Y'$-coordinate is almost parallel to the progressing direction of the eclipse-area on the duration considered here ($12^h 30^m$-$14^h 30^m$, or two hours centred at the maximum obscuration in the Japanese district). If we permit an assumption that eclipse effects at all points on the line parallel to the $Y'$-axis are similar to each other, we can determine magnetic forces corresponding to the eclipse effect at the cross-points of the rectangular mesh by the $X'$-$Y'$-coordinate-system, using the curves of four stations (Shimosato is omitted, as it is situated too close to Aso), and they are shown in Fig. 3A. To secure this figure, we applied the following consideration for each station: variations after the hour of the maximum obscuration represent the eclipse-effect at the rear-side (left, in this figure) of that station, and similary at the opposite side.

The current-system equivalent to the additional field due to the eclipse is derived from the horizontal vectors of this field. This field has a potential, but all these vectors do not satisfy this condition, as they introduce some errors. Then, in practice, the two figures corresponding to this field are drawn; one is derived from $X'$-component (3B) and the other from the $Y'$ (3C). As to the disagreement of those two figures, the method adopted by Hasegawa and Ota is introduced [2]. This method signifies the mean condition of the two figures, (3B) and (3C), and the result is shown in Fig. 3D. All Fig. 3's are the additional field due to the eclipse at the northern quarter of the eclipse-area, and the potential-difference of the consecutive line is $2.2 \times 10^3$ c.g.s., and the arrows show the direction of the current equivalent to the additional field. Next, the

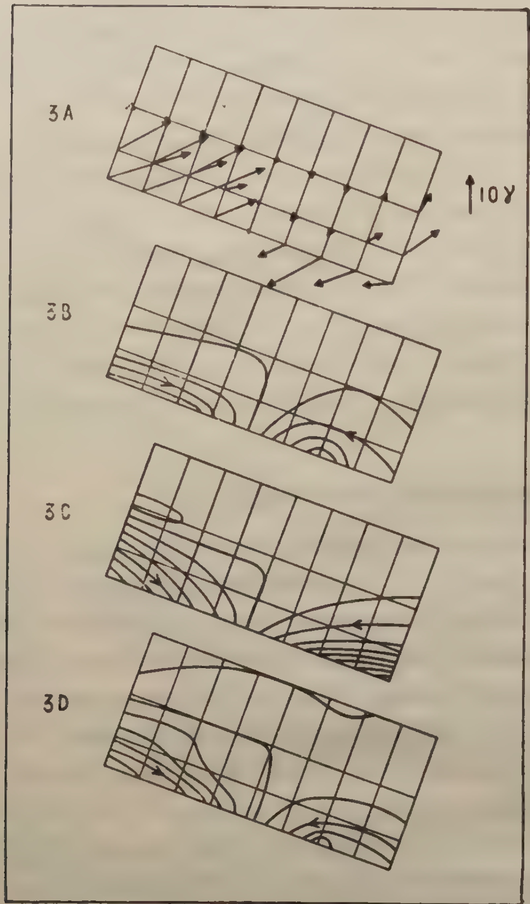


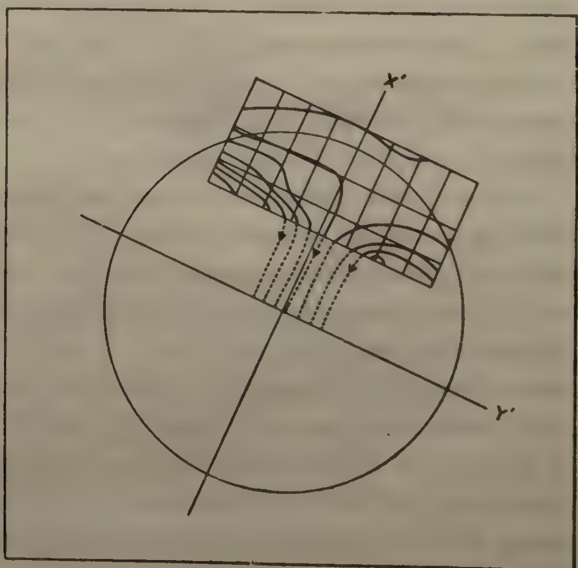

Fig. 3 Northern Quarter of Additional Field due to the eclipse.

additional field at the central part of the eclipse-area is considered. The general trend of the $S_q$-field at the central part of the present eclipse-area is not so simple as that of the northern quarter, as shown in Fig. 2. Then we utilize the previous result from the total eclipse, in which the diminution of the magnetic force was about half of the normal $S_q$ at that hour, and assume that the general trend of the $S_q$-field is spread in the same manner at the northern quarter. Fig. 4 shows the half-side of the additional field, obtained by combining the field thus obtained with the field shown in Fig. 3D. This result gives a typical example of the additional field due to ehe eclipse, under the condition of the uniform $S_q$-field.

## 4. Conclusion

The eclipse-effect depends on the condition of the variation field, and the affected area exceeds the eclipse-area, as the present result shows. It is difficult to draw the general picture of the

Fig· 4 Northern Half of Additional Field Potential Difference of consecutive lines is $2.2\times10^3$ c.g.s.



additional field due to the eclipse, as the variation field is too complex to be expressed by a simple model.

In conclusion, the writer wishes to express his sincere thanks to Prof. M. Hasegawa for his encouragement of this work, and also to Dr. A.K. Das, the Director General of Observatories sending us the observational data obtained in India.

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# The Instability of a Layer of Fluid Heated Below and Subject to the Simultaneous Action of a magnetic Field and Rotation

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(Read May 15, 1956; Received Aug. 15, 1956)

**Abstract**

Considering the instability of electrically cocducting fluid heated below when a uniform magnetic field and Coriolis acceleration are both present, we find the instability first sets in oscillation of increasing amplitude when a temperature gradient, however small, exists, under astrophysical condition $\varkappa \gg \frac{1}{4\pi\sigma} \gg \nu$ where $\varkappa$, $\sigma$ and $\nu$ are the thermal diffusivity, electrical conductivity and kinematic viscosity. Two critical frequency of oscillation are obtained depending on the ratio between Coriolis force and electromagnetic force.

## 1. Introduction

The effect of a magnetic field and of Coriolis acceleration on the thermal instability of a layer of fluid heated below has recently been investigated by Chandrasekhar ([1]—[7]).

The results revealed some very unexpected features. In this paper, we consider the condition for the occurrence of overstability when a uniform magnetic field and Coriolis acceleration are both present in the fluid of infinite conductivity and negligible kinematic viscosity.

## 2. The Equations of the problem

Consider a horizontal layer of fluid of depth $d$, confined between two parallel planes $z=0$ and $z=d$, subject to an effective gravity $\boldsymbol{g}$, acting in the direction of the vertical, a magnetic field $\boldsymbol{H}$ acting in a direction specified by a unit vector $\boldsymbol{\tau}$ and the Coriolis accerelation resulting from a rotation specified by a unit vector $\boldsymbol{\upsilon}$.

Expressing the prevailing magnetic field as the sum of two fields, the impressed field $\boldsymbol{H}$ (in the direction $\tau$) and a field $\boldsymbol{h}$ resulting from the motions, we start with the following equations, neglecting the terms $\nu\nabla^2 u_i$ and $\eta\nabla^2 h_i$ in Chandrasekhar's equations (cf. [7], eqns. (9), (10), (11) and (12)). All the notations are the same as in his equations.

$$\frac{\partial u_i}{\partial t}=-\frac{\partial \varpi}{\partial x_i}+\gamma\theta\lambda_i+2\Omega\epsilon_{ijk}u_j\nu_k+\frac{\mu H}{4\pi\rho_0}\tau_j\frac{\partial h_i}{\partial x_j}\ , \tag{1}$$

$$\frac{\partial h_i}{\partial t}=H\tau_j\frac{\partial u_i}{\partial x_j}\ , \tag{2}$$

$$\frac{\partial \theta}{\partial t}=\beta w+\kappa\nabla^2\theta\ , \tag{3}$$

$$\frac{\partial u_i}{\partial x_i}=\frac{\partial h_i}{\partial x_i}=0\ , \tag{4}$$

$$\varpi = \frac{p}{\rho_0} + \frac{\mu \boldsymbol{H}^2}{8\pi\rho_0} + g\lambda_i x_i - \frac{1}{2}\beta\gamma\lambda_i\lambda_j x_i x_j \quad , \tag{5}$$

where $\rho_0$ is a mean density, $w\,(=\lambda_j u_j)$ is the component of the velocity in the direction of the vertical, $\theta$ is the instantaneous departure of the temperature from the local mean value and $\gamma = g\alpha$, $\alpha$ being the coefficient of volume expansion of the fluid.

As is easily seen, we have no stationary solutions of the above equqtions. From these equations, we get

$$\frac{\partial \zeta}{\partial t} = 2\Omega\nu_j \frac{\partial w}{\partial x_j} + \frac{\mu H}{4\pi\rho_0}\tau_j \frac{\partial \phi}{\partial x_j} \quad , \tag{6}$$

$$\frac{\partial \chi}{\partial t} = H\tau_j \frac{\partial w}{\partial x_j} \quad , \tag{7}$$

$$\frac{\partial \phi}{\partial t} = H\tau_j \frac{\partial \zeta}{\partial x_j} \quad , \tag{8}$$

and

$$-\frac{\partial}{\partial t}\nabla^2 w = \gamma\left(\lambda_i\lambda_j \frac{\partial^2}{\partial x_i \partial x_j} - \nabla^2\right)\theta + 2\Omega\nu_j \frac{\partial \zeta}{\partial x_j} - \frac{\mu H}{4\pi\rho_0}\tau_j \frac{\partial}{\partial x_j}\nabla^2\chi \quad , \tag{9}$$

where $\zeta$, $\chi$ and $\phi$ are the $z$ components of the vorticity, magnetic filld $\boldsymbol{h}$ and curl $\boldsymbol{h}$ respectively.

Eliminating $\zeta$, $\phi$ $\chi$ and $\theta$ between these equations, we obtain

$$\begin{aligned} \left[\frac{\partial^2}{\partial t^2} - U^2\tau_i\tau_j \frac{\partial^2}{\partial x_i \partial x_j}\right]^2 \left[\frac{\partial}{\partial t} - \kappa\nabla^2\right]\nabla^2 w = \\ -\beta\gamma \frac{\partial}{\partial t}\left[\frac{\partial^2}{\partial t^2} - u^2\tau_i\tau_j \frac{\partial^2}{\partial x_i \partial x_j}\right]\left[\lambda_i\lambda_j \frac{\partial^2}{\partial x_i \partial x_j} - \nabla^2\right] w \\ -4\nabla^2 \frac{\partial^2}{\partial t^2}\nu_i\nu_j \frac{\partial^2}{\partial x_i \partial x_j}\left[\frac{\partial}{\partial t} - \kappa\nabla^2\right] w \quad , \end{aligned} \tag{10}$$

where $\qquad U = \dfrac{H}{\sqrt{4\pi\rho_0}}$ .

Equation (3), (6), (7), (8) and (9) represrnt the basic equations of this theory.

As the effect of viscosity is neglected, we must require

$$\theta = 0 \text{ and } w = 0 \text{ for } z = 0 \text{ and } z = d, \tag{11}$$

in either case the bounding surfaces are free or rigid.

From electromagnetic theory it follows that

$$E_x = E_y = 0 \text{ and } \chi = 0, \qquad \text{on a perfectly conducting surface.} \tag{12}$$

$$E_z = 0, \qquad \text{on a free surface adjoining a vacuum.} \tag{13}$$

### 3. The Case when $\boldsymbol{g}$, and $\boldsymbol{\Omega}$ act in the same Direction

When the axis of rotation ($\boldsymbol{\nu}$) coincides with the vertical, we may set

$$\boldsymbol{\lambda} = \boldsymbol{\nu} = (0,\ 0,\ 1),$$

and we take $\boldsymbol{\tau}$ in the direction of $\boldsymbol{y}$ axis

$$p_1 \sim \sqrt{2}\, p_0\,; \quad p_2 \sim \sqrt{2}\, \lambda p_0 \ . \qquad (34)$$

## 5. Concluding Remarks

From the preceeding section we see that under astrophysical conditions the instability first sets in as oscillations of increasing amplitude and the critical frequency differ from that of non-rotating fluid according to the ratio between the Coriolis force and the electromagnetic force. The value of $R$, when the instability of exponential increasing amplitude sets in, is determined only by the intensity of applied magnetic field $\boldsymbol{H}$ and the density of fluid $\rho_0$, independent of angular velocity $\boldsymbol{\Omega}$.

## 6. Acknowledgement

The auter wishes to express his thanks to Prof. M. Hasegawa for his constant interest in and encouragements.

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**Meeting of the Society of Terrestrial Magnetism and Electricity:**

The 19th General Meeting was held at the Scientific Research Institute on May 14—16, 1956.

Number of the reports read at the Meeting;

Rock Magnetism, 8; Cosmic Rays, 12; Geomagnetism and Earth Current, 13; Ionosphere, 14; Atmospheric Electricity and Atmospherics, 15.

Tanakadate Prize was awarded for the following excellent workers:

The 19th, Mr. I. Yokoyama;

Geomagnetic Studies of Volcano Mihara.

昭和31年9月20日 印刷
昭和31年9月25日 發行
第8巻 第2號

編輯兼發行者 日本地球電氣磁氣學會
代表者 長谷川万吉

印刷者 京都市南区上鳥羽唐戸町63
田中幾治郎

賣捌所 丸善株式會社京都支店
丸善株式會社 東京・大阪・名古屋・仙台・福岡

JOURNAL OF GEOMAGNETISM AND GEOELECTRICITY

Vol. VIII No. 2

1 9 5 6

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